Soot particulate formation and characterization in combustion
Gulder, Omer L.

National Research Council Canada   Conseil national de recherches Canada

Canadä

SOOT PARTICULATE FORMATION AND CHARACTERIZATION IN COMBUSTION

Ömer L. Gülder

Combustion Research Group
ICPET, Bldg. M-9
National Research Council Canada
Ottawa, Ontario K1A 0R6

ABSTRACT

The subject of soot formation in combustion is being extensively studied to decipher the relevant mechanisms. Despite considerable progress made in understanding some of the features of soot formation, we still do not have sufficient details of the formation mechanisms to be able to calculate soot in the flame of a given hydrocarbon fuel from first principles. This paper first gives an overview of the soot particulate formation in combustion and its contribution to ambient particulate matter. The experimental burner types that are commonly used for soot research are then reviewed and basic features of the most commonly used laminar diffusion burners are summarized. Soot particulate formation and emissions in diesel and gasoline engines, and their relative contributions to ambient particulate matter, are discussed. Methods based on physical sampling and non-intrusive laser-based soot diagnostic techniques are briefly reviewed and their capabilities and limitations are discussed.

1. INTRODUCTION

A significant portion of atmospheric particulates arises from combustion of fuels in various engines and furnaces. In urban areas, mobile sources are major contributors to ambient particulate matter (PM) concentrations. The main constituent of the particulates generated by combustion is carbon. These carbonaceous particulates, which are produced from gas-phase combustion processes, are generally called soot, and those that form as a result of direct pyrolysis of liquid hydrocarbon fuels are generally referred to as coke or cenospheres.

The detrimental effect of soot particulates on human health is a current concern and various restrictions are being placed on particulate matter emissions from vehicles and other sources. From an operational point of view, soot formation is not desired in most power plants. For example, in gas turbines, the presence of excessive soot reduces the combustor liner life as well as contributing to the mechanical erosion of turbine blades and nozzle guide vanes. In military applications visible exhaust plumes enhanced by the presence of soot particulates are highly undesirable. On the other hand, in industrial furnaces of relatively large capacities, the major portion of heat transmission is through thermal radiation, mainly facilitated by the presence of soot particles in the combustion volume. In the absence of soot particulates a substantial portion of the released energy from the fuel, instead of being transmitted directly to the heat absorbing working fluid by radiation, must be appropriately extracted by convection leading to larger and more expensive systems capable of handling high temperature gases. The challenge in these furnaces is to boost radiative heat transfer by promoting soot production rate in such a manner that the soot is completely oxidized before leaving the combustion volume.

To be able to control soot in combustion, a detailed understanding of the key features of its formation and oxidation is required. Although considerable progress has been made in recent years in the understanding of the soot particulate formation in combustion, the essential details of the formation mechanisms are sketchy at best. The state-of-the-art is not yet at a stage where one could describe the process of soot formation from a selected chemical compound under well-defined conditions.

2. AMBIENT PARTICULATE MATTER

There is a mixture of pollutants in ambient air classified as particles or particulate matter. They may form in the atmosphere or originate from a variety of sources. Their structure, morphology, size, and composition depend on the source. There is a substantial contribution from fossil fuel combustion to ambient

particulates of respirable size. The mass emissions rate of particulates from on-road diesel vehicles has already been regulated, and to meet the increasingly stringent limits, the manufacturers have continually improved engine designs and incorporated exhaust control devices. Although health professionals suspect that the size, number density, and chemical composition of the particles are more relevant to health effects than concentration, these parameters are neither regulated nor routinely measured.

The total suspended particulates in ambient air are crudely classified as follows [1]:
a. nonrespirable fraction (50 to 100 μm)
b. PM_{10} (smaller than 10 μm)
c. $PM_{2.5}$ (smaller than 2.5 μm)
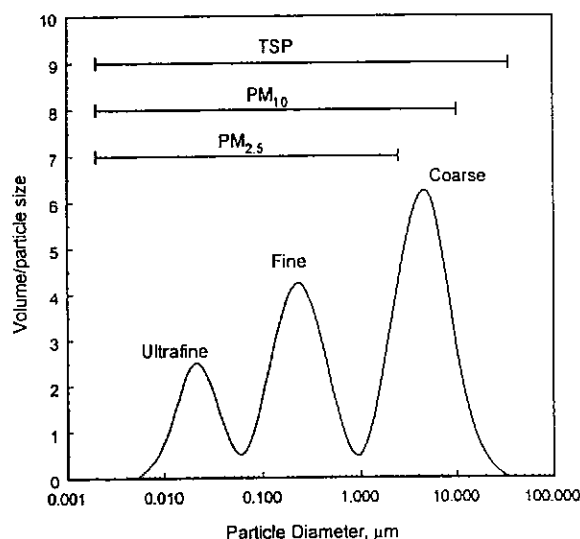d. $PM_{0.1}$ (smaller than 0.1 μm)

Figure 1. A typical volume-weighted size distribution of ambient particulates. TSP = total suspended particulates. Adopted from [1].

Total particulates tend to fall into three modes as seen in Fig.1. Since most particulates in ambient air are not spherical, the size is usually designated by the aerodynamic diameter. Figure 1 shows a typical volume-weighted size distribution with three commonly used measures of ambient particle concentration, which are based on the size cutpoint of the sampling and measurement systems. Particles from on-road vehicles are primarily found in the ultrafine and fine modes. The exhaust soot particulate from on-road vehicles may contribute to street level particulate matter in urban centres by up to 50%.

The most widely used ambient particulate matter sampling techniques are:

- Dichotomous sampler: low-volume manual sampler, collects fine and coarse fractions of PM_{10} on Teflon membrane filters.

- Size Selective High Volume Sampler: this is a high-volume manual sampler, collects PM_{10} on Teflon coated or quartz fibre filters.

- Tapered Element Oscillating Micro-balance (trade name is TEOM): an automated instrument giving a continuous record of mass of PM_{10} or $PM_{2.5}$ depending on the inlet used.

- Partisol Sampler: low-volume manual sampler, collects either PM_{10} or $PM_{2.5}$ on Teflon filters; reference method for $PM_{2.5}$.

3. SOOT PARTICULATE FORMATION IN COMBUSTION

The formation of soot particulate, i.e. the conversion of a hydrocarbon fuel with molecules containing a few carbon atoms into a carbonaceous agglomerate containing some millions of carbon atoms in a few milliseconds, is an extremely complex process. This process involves a transition from gaseous to solid phase where the solid phase does not exhibit any unique chemical and physical structure. The accepted mechanism for this process is as follows (see, e.g. [2-5]): The hydrocarbon fuel goes through pyrolysis or partial oxidation during combustion and forms small hydrocarbon radicals from which small hydrocarbons, particularly acetylene(s), are formed. Reaction of C_4 species (e.g. diacetylene) with C_2 species (acetylene) produce a C_6 species which can form an aromatic structure through isomerization. It is also possible to have C_3H_3 dimerization with subsequent isomerization to an aromatic structure. The formation of larger aromatic rings occurs mainly via an acetylene addition mechanism. Coagulation of these larger aromatic ring compounds is proposed to account for the formation of primary soot particles. The smallest of these particles, when observed under the electron microscope, are about 1.5 nm in diameter, corresponding to a molecular mass of about 2000. These primary particles then pick up molecules from the gas phase promoting surface growth, whose rate is one of the determining factors in final soot concentration. The final size of the soot particles results from coagulation of primary particles to larger aggregates. A schematic representation is given in Fig.2.
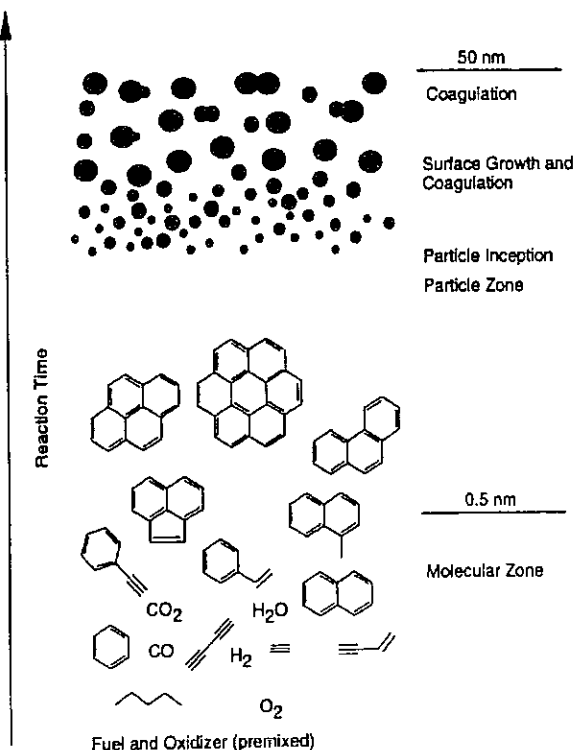
Figure 2. A schematic representation of soot formation under premixed combustion conditions [2].

Figure 3. A smoke point laminar diffusion flame in the author's laboratory.
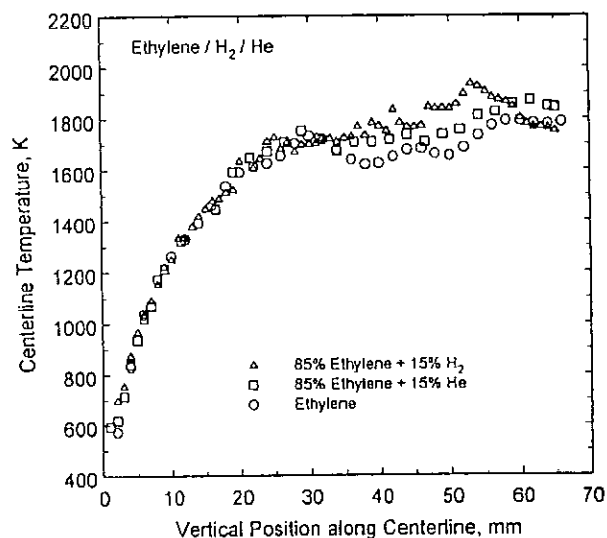
Figure 4. Measured centerline temperature profiles as a function of axial position along the flame centerline in ethylene flames with and without hydrogen or helium dilution [6]. Measurements taken by CARS on the burner shown in Fig.3.

Diffusion Flames

Turbulent diffusion flames are the mode of combustion in most practical combustion devices, including diesel engines. Soot formation is an artifact of this mode of combustion. In premixed flames, unless the fuel-air ratio is very rich, soot does not form. The non-homogeneous nature of turbulent diffusion flames makes it very difficult to isolate parameters that affect soot formation and oxidation. For this reason the more easily controlled laboratory experiments are performed in shock tubes and laminar diffusion flames. One disadvantage of the shock tubes is that they have a very short residence time as compared to practical flames. There are three types of laminar diffusion flame burners that are commonly used by soot researchers: the coflow, counterflow, and the Wolfhard-Parker burners. Although the counterflow (opposed jet) and the Wolfhard-Parker burners produce almost one-dimensional flames, they may suffer from stability problems under certain flow conditions. In addition, in the counterflow burner flames the stagnation point location is very critical and it can vary by the choice of flow conditions. Coflow burner flames are radially axisymmetric two-dimensional flames with good stability. Most of the

research work on soot formation and oxidation has been conducted on coflow laminar diffusion flames.

A widely used coflow laminar diffusion flame burner consists of a fuel nozzle made of 12.7 mm diameter stainless-steel pipe providing the gaseous fuel. Air is supplied from a concentric converging nozzle of about 100 mm diameter. The flame generated re-

3

Figure 5. Measured temperature profiles as a function of radial position at two axial heights in ethylene flames with and without hydrogen or helium dilution [6]. Measurements taken by CARS on the burner shown in Fig.3.

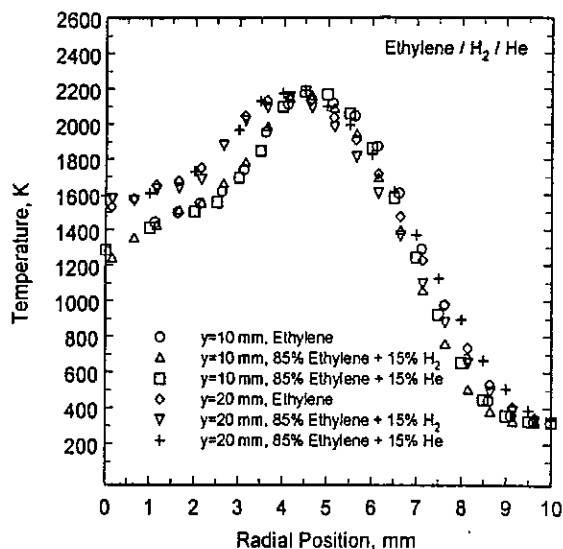sembles that of a candle flame (Fig.3). Coflow laminar diffusion flames are buoyancy dominated and the flame height is proportional to the fuel flow rate. The smoke point is the maximum fuel flow rate at which no soot escapes from the tip of the flame. Figure 3 shows an ethylene diffusion flame burning just below its smoke point height. Temperature measurements in this flame obtained by CARS (coherent anti-Stokes Raman spectroscopy) are shown in Figures 4 and 5 [6]. It has proved impossible to measure accurately temperatures in these flames using conventional thermocouple techniques. Such data are required to decipher the effect of flame temperature on soot formation.

Figure 6 shows a schematic picture constructed from the soot morphological data obtained by the thermophoretic sampling technique combined with the laser light scattering and extinction measurements in a coflow laminar ethylene diffusion flame. Micrographs obtained by transmission electron microscopy from the soot samples at two different locations, one at soot formation and growth region and the other at where soot growth is completed, are shown in Figures 7 and 8.

Note that in the growth region, both the primary particle size and the number of primary particles in the agglomerate are both increasing (Fig.6). In the oxidation region, the size of the primary particles is
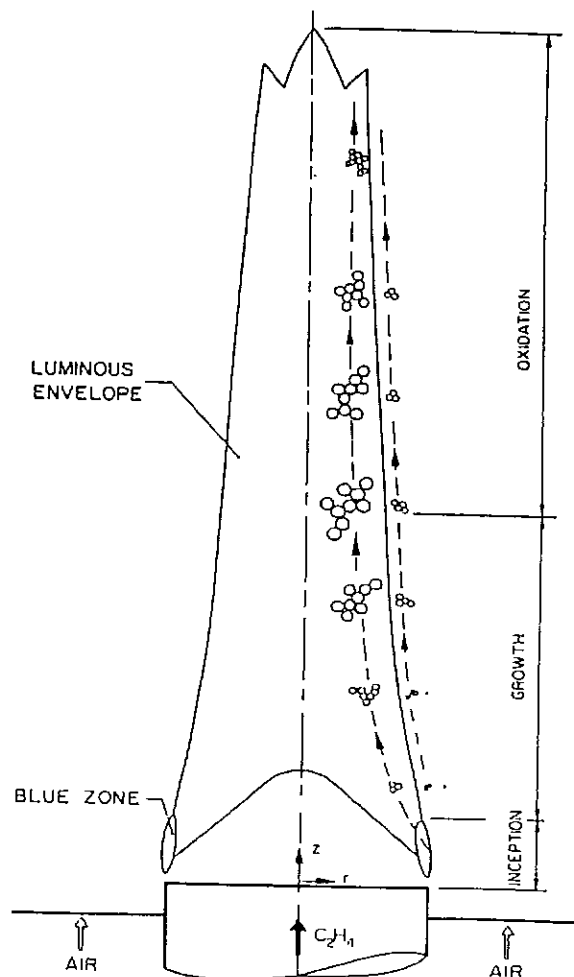
Figure 6. A schematic representation of soot aggregate morphological evolution in the coflow ethylene laminar diffusion flame. The particle path inside the luminous envelope corresponds to the location of the maximum soot volume fraction, while the one on the outside corresponds to location closer to the flame sheet [7].

reduced while the number of particles in the agglomerate remains unchanged.

Figure 9 shows the primary soot particle size in two coflow laminar diffusion flames as a function of flame height along the streamline of maximum soot concentration. The lower fuel flow rate flame is a smoke point flame in which the soot formed is completely oxidized within the luminous visible flame. In the high fuel flow flame, soot particles are not completely oxidized and they escape from the tip of the flame (smoking flame). It seems that the size of the particles are the same up to the flame height of 50 mm. In the case of the smoking flame, a reduced

Figure 7. Micrograph of soot particles sampled from a height of 10 mm and a radial location of 5 mm in the flame shown in Fig.6 [8].

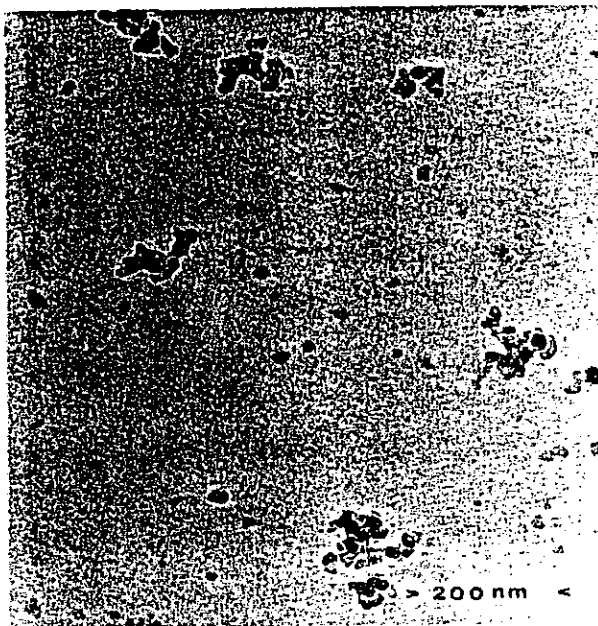Figure 9. Primary soot particle diameter as a function of flame height along the maximum soot concentration streamline in two ethylene-air laminar diffusion flames. Data adopted from Megaridis and Dobbins [7,9].

and nearly spherical primary particles [7,9]. The aggregates vary in the number of primary particles they contain, which is often described by a log-normal distribution.

The important parameters that influence the formation of soot in such flames are the parent fuel molecular structure [4,10,11], the flame temperature [12-15], the amount and nature of diluents either in the fuel or in the oxidizer [6,13,15-20], and the pressure [21]. In practical combustion devices, the situation is more complex due to turbulent flow (two-phase in most systems), complex fuel chemistry of distillate fuels, and the operating conditions, especially transients.

Premixed Flames

Premixed burners used in soot research offer one-dimensional stable flames which lend themselves for extensive intrusive and non-intrusive measurements. In low pressure premixed flames, the reaction zone gets thicker which provides excellent spatial resolution (which translates into a good temporal resolution). Most of the mass spectroscopy measurements (including time-of-flight) have been done on premixed flames, see e.g. [22-24]. Soot formation in rich premixed flames more or less follows the same route as in diffusion flames, except that the situa-
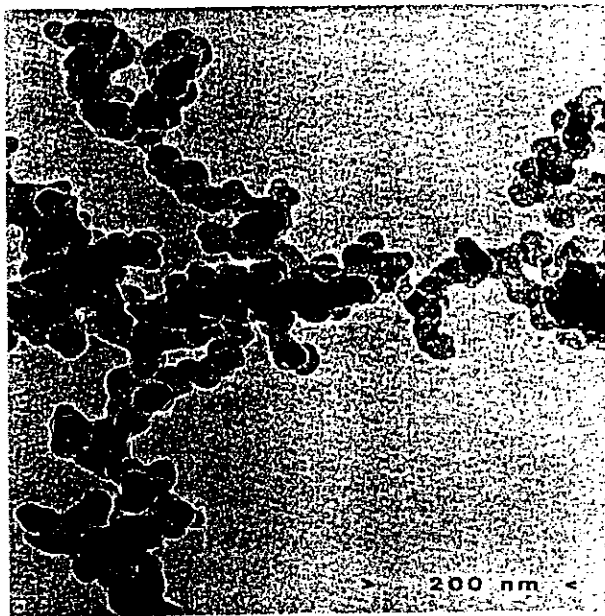

Figure 8. Micrograph of soot particles sampled from a height of 330 mm on the centerline in the flame shown in Fig.6 [8].

oxidation rate allows particles to escape after partial oxidation. Thermophoretic sampling and transmission electron microscopy examination of the soot from this type of flame have shown that the soot particles consist of aggregates of nearly monodisperse
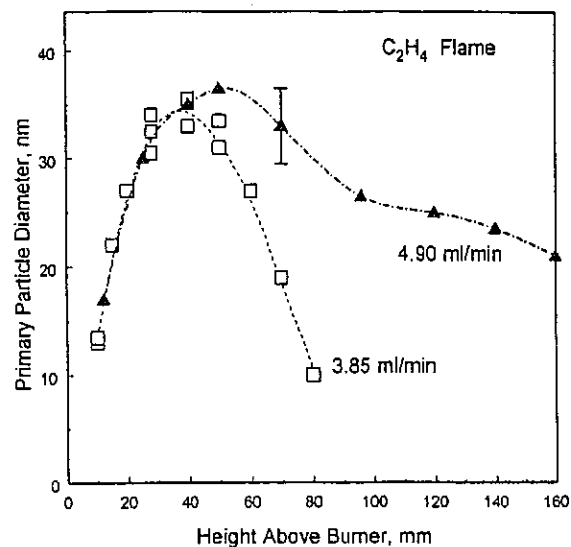
5

tion is not complicated by the presence of fuel and oxidizer mixing found in diffusion flames.

Figure 2 shows a general schematic picture of soot formation under premixed conditions. Soot formation processes under premixed combustion conditions are not expected to be influenced by the flame turbulence [4]. Results from turbulent bunsen burners and stirred reactors seem to support this contention.

The relative tendency of fuels to form soot under premixed conditions generally has been measured by observing the critical equivalence ratio, or the C/O ratio, at which soot just forms [25]. It has been shown that the critical equivalence ratios depend on the flame temperature [25,26] and the OH (hydroxyl radical) concentration [27]. In premixed flames the higher the flame temperature, the less the tendency to soot. This trend, which is the just opposite of the diffusion flames, has been attributed to the fact that as the temperature rises, oxidative attack on soot precursors increases at a faster rate than precursor formation. Takahashi and Glassman [25] have shown that at a fixed flame temperature the critical equivalence ratio could correlate with the "number of C-C bonds" in the fuel. It seems that, under premixed conditions, all fuels must break down to the same essential species–postulated to be mostly acetylene [25]–which builds into a soot particle. This seems to be supported by the findings of Harris and Weiner [24].

In most premixed combustion applications, the operating equivalence ratio is much leaner than the critical equivalence ratio and hence no soot particulate formation is expected in such flames. However, in the presence of nonhomogeneous pockets, the local equivalence ratio may be rich enough to form soot, although the overall equivalence ratio is leaner than the critical value. One example of this situation is the cold start conditions in a fuel injected gasoline engine in which the local equivalence ratios may exceed the critical values for soot formation.

Particulate Emissions from Diesel Engines

The particulate emissions from diesel engines are in the form of complex aerosols consisting primarily of soot and volatile organics. There is limited information currently available on the size and morphology of diesel particulates; most measurements have been for concentration only. However, soot itself has been studied in more detail. Most soot particles are agglomerates of 5 to 30 nm diameter primary particles. Typical dimensions of these agglomerates

range from 10 to 1000 nm, but most of the mass is log-normally distributed with mass median diameters on the order of 100 to 250 nm [28,29]. These dimensions of the diesel soot particles are very similar to those obtained from atmospheric diffusion flames of gaseous hydrocarbon fuels (Figures 7-9). As the particulate concentrations in the exhaust of modern diesel engines are reduced, the agglomerate sizes are also being decreased.

Figure 10 shows the average composition of the typical diesel exhaust particulate matter. Inorganic components in the diesel exhaust are products of engine and component wear, or are trace contaminants of the fuel and the lubricant. The metals found in the particulate matter are primarily trace fuel contaminants such as antimony, arsenic, barium, beryllium, cobalt, and strontium. These substances will vaporize in the combustion chamber and then stick to particles in the exhaust stream.
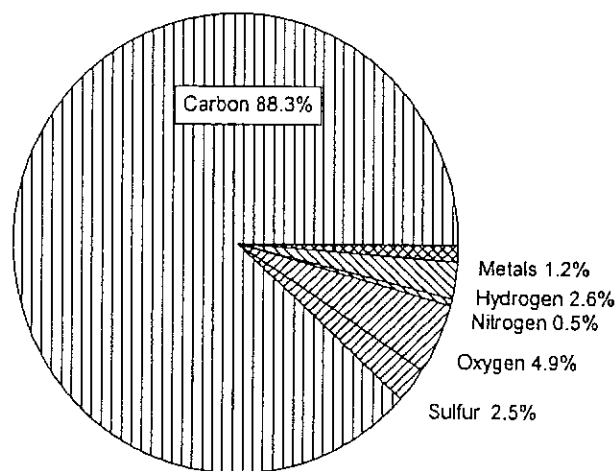
Figure 10. Elemental composition of the typical diesel particulate matter by mass [31].

For regulatory purposes, particulate matter emissions are defined as the mass of the matter that can be collected from a diluted exhaust stream on a filter kept at 52 °C. This includes the organic compounds that condense at lower temperatures, but excludes the condensed water. Exhaust gas temperature, before the exhaust is diluted, plays an important role on the amount of adsorbed soluble organics; low exhaust gas temperatures produce PM with more soluble organics than PM measured from a high temperature exhaust [30].

Particulate Emissions from Gasoline Engines

There is limited information on the size, morphology, and concentration of the particulate emissions from gasoline engines. The particulate size is much finer than the diesel particulates and the concentrations are below the accurate detection limit of standard detection techniques [1,32,33]. The majority of the particles from gasoline engine exhaust have aerodynamic diameters in the range of 20 to 200 nm, with a peak at about 50 nm [1]. The particulate emissions from gasoline engines are not currently legislated. However, the adverse health effects of submicron particulates have been recognized by EPA and the other legislating bodies. It should be noted that gasoline engines may contribute up to 25% of the transportation generated particulate matter in urban areas.

The source and the mechanism of particulate formation in gasoline engines are largely unknown. In a recent study [34], it has been shown that, under steady state engine running conditions, the particulate matter comprise mostly of soot particles from combustion when the mixture is fuel rich (fuel-air equivalence ratio larger than about 1.43) . However, when the mixture is in the lean to slightly rich region (fuel-air equivalence ratio between 0.83 and 1.25), the particulate matter is mostly derived from the lubricating oil.

4. MEASUREMENT OF SOOT PARTICULATES

Exhaust Particulate Measurements

For regulatory purposes, the particulate matter in the exhaust is measured by collecting the PM on a filter in a dilution tunnel. This measurement gives the averaged PM emissions over the time period during which the particulates are collected on the filter. For exhausts containing lower concentrations of PM, the collection time could be very long for collecting enough PM for reasonable measurements. In spite of its drawbacks and limitations, the filter technique is being used for diesel engine and fuel certification and testing. Since the collected PM and other condensed material on the filter stick together, it is almost impractical to determine the particulate size and size distribution. However, the soluble organic fraction can be extracted from the collected material and analyzed for its chemical composition.

One of the principal instruments for particulate size measurements for particulates within an aero-

dynamic diameter range of 0.01 to 1 μm has been the differential mobility particle sizer (DMPS). This device separates charged particles according to their aerodynamic size using an electric field. Separated particles are then counted using a condensation nucleus counter which yields a number-weighted size distribution. Since the response time of the instrument is on the order of a minute, it is not suitable for transient measurements [1].

A fairly new instrument capable of measuring particle sizes in the 0.04 to 10 μm range is the electrical low-pressure impactor (ELPI). Particulates are first charged and then passed through a cascade impactor, which segregates them into a number of bins (size ranges). Changes in the current deposited on each impactor stage yield the transient concentrations of particles of the corresponding aerodynamic size [1].

Non-Intrusive Soot Measurements

In combustion research soot volume fraction measurements are important for studies of soot formation, radiation processes, and for monitoring post-flame particulates. Light extinction is a commonly used diagnostic technique for measuring soot volume fraction. However, it suffers from the drawback of measuring a line-of-sight average, and while tomographic reconstruction can be used to calculate soot profiles in radially symmetric flames, this is not possible in turbulent flames. Elastic scattering of light has been widely investigated for soot measurements but the fact that the signal is proportional to the square of the particle diameter means that the technique is more useful for particle sizing than volume fraction measurements. More importantly, for agglomerated soot particles (which are definitely not spherical), it has become increasingly clear in the last few years that the approach of applying Mie theory by assuming spherical soot particles results in large errors [35-38].

By combining the elastic scattering and extinction techniques it is possible to obtain the soot particulate morphological parameters. Köylü [39] describes an in situ particulate diagnostic technique based on the Rayleigh-Debye-Gans polydisperse fractal aggregate scattering interpretation of absolute angular light scattering and extinction measurements. Using the proper particle refractive index, the method yields particle volume fraction, fractal dimension, primary particle diameter, particle number density, and aggregate size distribution [39]. However, the technique is complex (the absolute scattering cross-section must be measured as a function of scattering angle) and is

limited to laminar cases (laminar flames and laminar gas flows) and cannot be used in turbulent environments.

Laser induced incandescence (LII) has emerged as a promising technique for measuring spatially and temporally resolved soot volume fraction in flames [40-49]. In LII the soot is heated by a short duration laser pulse to produce incandescence. With sufficiently high laser energies, numerical models of the heat transfer indicate that the soot particles reach temperatures of 4000-4500 K [40,41,45,47,48]. The resultant radiation, which is blue shifted relative to soot radiation at normal flame temperatures and is of short duration, can readily be detected. LII typically has a temporal resolution of 10 ns and can be used to perform both point measurements and 2-D planar visualization. The schematic of the LII setup at NRC is shown in Figure 11.
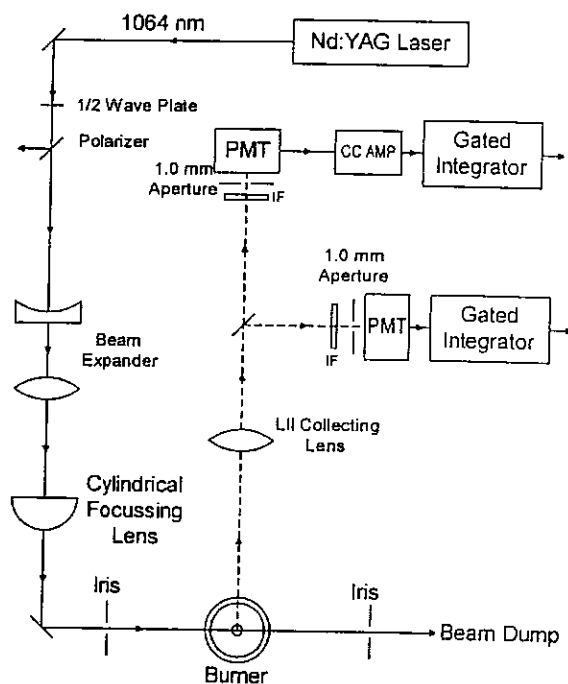

Figure 11. Schematic diagram of LII layout [50].

LII uses a pulsed focused laser beam to provide an instantaneous (about 10 ns) energy source. Several mJ of energy is used to rapidly heat particulates to their evaporation temperature. The soot particulates radiate incandescence as they cool back to the ambient temperature which is about 1500 to 2000 K in flames. The incandescence signal is collected and imaged onto a detector as a function of time. This signal is proportional to the particulate volume fraction over a very wide dynamic range and it is also

possible to determine the particulate size [50]. It can be used in any environment, including laminar and turbulent flames and exhaust gas streams. Figure 12 shows a comparison of measurements of soot volume fraction in a laminar diffusion flame by LII and by line-of-sight extinction.
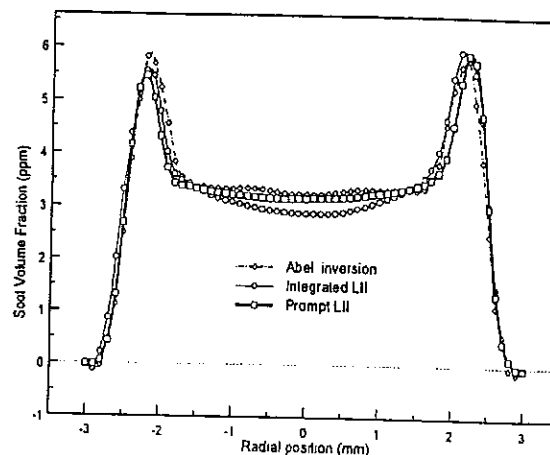

Figure 12. Comparison of soot volume fraction determined by LII to radial profiles from Abel inversion of line-of-sight extinction data at 40 mm above the burner in a laminar diffusion flame of ethylene and air [50].

5. CONCLUDING REMARKS

The details of the chemical and physical mechanisms of soot formation process in combustion remain uncertain due to the highly complex nature of hydrocarbon flames. The mechanisms of soot formation have been investigated extensively over many years and only very broad features of the formation processes have been established. In addition to great uncertainties regarding formation mechanisms, the control and prediction of soot levels in practical combustion devices are further impeded by the complexities introduced by the transient operating conditions, existence of a two-phase turbulent flow field, and the chemistry of distillate fuels consisting of several hundred hydrocarbon compounds.

The hope of reducing or eliminating soot particulate emission from combustion sources clearly requires a better understanding of its formation and oxidation during the combustion process. The level of understanding required to fully control soot may not be available for some time because of the complexity of the process. However, the advancement of laser-based combustion diagnostic techniques for measur-

8

ing spatially and temporally resolved soot morphology, species concentrations, and flame temperature combined with detail chemical kinetic modelling efforts contribute to deciphering the complex issue of soot formation.

The complex nature of engine combustion makes the diesel engine unsuitable for more fundamental studies. It is very difficult to interpret fundamental ideas regarding soot formation in the engine combustion context. However, a basic understanding of soot formation and oxidation is central to the control of particulate emissions from combustion sources. One- and two-dimensional laminar flames are suitable devices to obtain this basic understanding at atmospheric conditions. Due to the very short time scales associated with various stages of soot formation and oxidation, only simple, but well-controlled, laboratory flames can provide the detailed information required.

6. REFERENCES

[1] Anon., 1997, "Formation and Characterization of Particles: Report of the 1996 HEI Workshop", Health Effects Institute HEI Communications, Number 5, September.

[2] H. Bockhorn(Ed.), 1994, *Soot Formation in Combustion*, Springer-Verlag, Berlin.

[3] I. Glassman, 1988, "Soot Formation in Combustion Processes", 22nd Symposium (International) on Combustion, The Combustion Institute, p.295-310.

[4] I. Glassman, 1996, *Combustion*, 3rd Edition, Academic Press, San Diego, Chap.8.

[5] M. Frenklach and H. Wang, 1994, "Detailed Mechanism and Modelling of Soot Particle Formation" in *Soot Formation in Combustion* (Bockhorn, H., (Ed.)), Springer-Verlag, Berlin, pp.165-192.

[6] Ö. L. Gülder, D. R. Snelling and R. A. Sawchuk, 1996 "Influence of Hydrogen Addition to Fuel on Temperature Field and Soot Formation in Diffusion Flames", 26th Symposium (International) on Combustion, The Combustion Institute, pp.2351-2357.

[7] C.M.Megaridis and R.A.Dobbins, 1988, "Soot Aerosol Dynamics in A Laminar Ethylene Diffusion Flame", 22nd Symposium (International) on Combustion, The Combustion Institute, pp.353-362.

[8] R. A. Dobbins and H. Subramaniasivam, 1994, "Soot Precursor Particles in Flames" in *Soot Formation in Combustion* (Bockhorn, H., (Ed.)), Springer-Verlag, Berlin, pp.290-299.

[9] C. M. Megaridis and R. A. Dobbins. 1989, "Comparison of Soot Growth and Oxidation in Smoking and Non-Smoking Ethylene Diffusion Flames", Combust. Sci. Technol. 66, pp.1-16.

[10] Ö. L. Gülder, 1989, " Influence of Hydrocarbon Fuel Structural Constitution and Flame Temperature on Soot Formation in Laminar Diffusion Flames", Combust. Flame, 78, pp. 179-194.

[11] Ö. L. Gülder and B. Glavincevski, 1991 "Influence of Fuel-Bound Sulfur on Soot Formation in Laminar Diffusion Flames of Liquid Hydrocarbons", Combust. Sci. Technol. 77, pp. 337 - 343.

[12] R. L. Axelbaum, W. L. Flower and C. K. Law, 1988, "Dilution and Temperature Effects of Inert Addition on Soot Formation in Counterflow Diffusion Flames", Combust. Sci. Technol. 61, pp.51-73.

[13] D. X. Du, R. L. Axelbaum and C. K. Law, 1991, "Experiments on the Sooting Limits of Aerodynamically Strained Diffusion Flames ", 22nd Symposium (International) on Combustion, The Combustion Institute, pp.387-394.

[14] Ö. L. Gülder, 1992, "Soot Formation in Laminar Diffusion Flames at Elevated Temperatures", Combust. Flame, Vol.88, pp.74-82.

[15] Ö. L. Gülder and Snelling, D. R., 1993, "Flame Temperature and Dilution Effects on Soot Formation in Diffusion Flames", Combust Flame, 92, pp.115 - 124.

[16] H. S. Hura and I. Glassman, 1988, "Soot Formation in Diffusion Flames of Fuel / Oxygen Mixtures", 22nd Symposium (International) on Combustion, The Combustion Institute, pp.371-378.

[17] C. Wey, E. A. Powell and J. I. Jagoda, 1984, "The Effect on Soot Formation of Oxygen in the Fuel of a Diffusion Flame", 20th Symposium (International) on Combustion, The Combustion Institute, pp.1017-1024.

[18] K. P. Schug, Y. Manheimer-Timnat, P. Yaccarino and I. Glassman, 1980, "Sooting Behaviour of Gaseous Hydrocarbon Diffusion Flames and the Influence of Additives", Combust. Sci. Technol. 22, pp.235-250.

[19] Ö. L. Gülder, 1993, "Influence of Sulphur Dioxide on Soot Formation in Diffusion Flames", Combust. Flame, 92, pp.410 - 418.

[20] Ö. L. Gülder, 1995, "Effects of Oxygen on Soot Formation in Methane, Propane, and n-Butane Diffusion Flames", Combust. Flame, 101, pp. 302-310.

[21] W. L. Flower and C. T. Bowman, 1986, "Soot Production in Axisymmetric Laminar Diffusion Flames at Pressures from One to Ten Atmospheres", 21st Symposium (International) on Combustion, The Combustion Institute, pp. 1115-1124.

[22] B.L.Wersborg, J.B.Howard and G.C.Williams, 1974, "Physical Mechanism in Carbon Formation in Flames", 14th Symposium (International) on Combustion, The Combustion Institute, pp.929-940.

[23] J. T. McKinnon and J. B. Howard, 1992, "The Role of PAH and Acetylene in Soot Nucleation and Growth", 24th Symposium (International) on Combustion, The Combustion Institute, pp.965-971.

[24] S. J. Harris and A. M. Weiner, 1984, "Some Constraints on Soot Particle Inception in Premixed Ethylene Flames", 20th Symposium (International) on Combustion, The Combustion Institute, pp.969-978.

[25] F. Takahashi and I. Glassman, 1984, "Sooting Correlations for Premixed Flames", Combust. Sci. Technol. 37, pp. 1-19.

[26] H. F. Calcote and D. B. Olson, 1982, "Importance of Temperature on Soot Formation in Premixed Flames", Combust. Sci. Technol. 28, pp.315-317.

[27] M. M. Harris, G. B. King and N. M. Laurendeau, 1986, "Influence of Temperature and Hydroxyl Concentration on Incipient Soot Formation in Premixed Flames", Combust. Flame, 64, pp.99-112.

[28] D. F. Dolan, D. B. Kittelson and D. Y. H. Pui, 1980, "Diesel Exhaust Particle Size Distribution Measurement Techniques", SAE Paper No. 800187, Society of Automotive Engineers, Warrendale, PA.

[29] R. L. Williams, R. L., 1982, "Diesel Particulate Emissions", in *Toxicological Effects of Emissions from Diesel Engines*, (J. Lewtas (Ed.)), Elsevier, New York, pp.15-32.

[30] Y. Kishi, H. Tohno and M. Ara, 1992, "Characteristics and Combustibility of Particulate Matter", in *Reducing Emissions from Diesel Combustion*, Society of Automotive Engineers, Warrendale, PA. pp. 139-146.

[31] Anon., 1989, "Unregulated Motor Vehicle Exhaust Gas Components", Volkswagen AG, Research and Development (Physico-Chemical Metrology). Project coordinator: K. H. Lies. 3180 Wolfsburg 1, Germany.

[32] S. J. Greenwood, J. E. Coxon, T. Bidduiph and J. Bennet, 1996, "An Investigation to Determine the Exhaust Particulate Size Distributions for Diesel, Petrol, and Compressed Natural Gas Fueled Vehicles", SAE Paper No. 961085, Society of Automotive Engineers, Warrendale, PA.

[33] D. J. Rickeard, J. R. Bateman, Y. K. Kwon, J. J. McAughey and C. J. Dickens, 1996, "Exhaust Particulate Size Distribution: Vehicle and Fuel Influences in Light Duty Vehicles", SAE Paper No.961980, Society of Automotive Engineers, Warrendale, PA.

[34] Y. Shin and W. K. Cheng, 1997, "Engine-Out Dry Particular Matter Emissions from SI Engines", SAE Paper No. 972890, Society of Automotive Engineers, Warrendale, PA.

[35] R. A. Dobbins and C. M. Megaridis, 1987, "Morphology of Flame-Generated Soot as Determined by Thermophoretic Sampling", Langmuir, 3, pp.254-259.

[36] R. A. Dobbins and C. M. Megaridis, 1991, "Absorption and Scattering of Light by Polydisperse Aggregates", Appl. Optics, 30,pp.4747-4754.

[37] Ü. Ö. Köylü and G. M. Faeth, 1994, "Optical Properties of Overfire Soot in Buoyant Turbulent Diffusion Flames at Long Residence Times",J. Heat Trans., 116, pp.152-159.

[38] Ü. Ö. Köylü and G. M. Faeth, 1994, "Optical Properties of Soot in Buoyant Laminar Diffusion Flames," J. Heat Trans. 116, pp.971-979.

[39] Ü. Ö. Köylü, 1996, "Quantitative Analysis of In Situ Optical Diagnostics for Inferring Particle / Aggregate Parameters in Flames: Implications for Soot Surface Growth and Total Emissivity", Combust. Flame, 109, pp.488-500.

[40] C. J. Dasch,1984, "New Soot Diagnostics in Flames Based on Laser Vaporization of Soot", 20th Symposium (International) on Combustion, The Combustion Institute, pp.1231-1237.

[41] L. A. Melton,1984, "Soot Diagnostics Based on Laser Heating", Appl. Optics, 23, pp.2201-2208.

[42] J. E. Dec, A. O. zur Loye and D. L. Siebers,1991, "Soot Distribution in a D.I. Diesel Engine Using 2-D Laser Induced Incandescence Imaging", SAE Transactions, 100, pp.277-288.

[43] R. L. Vander Wal and K. J. Weiland, 1994, "Laser-Induced Incandescence: Development and Characterization Towards a Measurement of Soot Volume Fraction", Appl. Phys. B59, pp.445-452.

[44] R. Puri, T. F. Richardson, R. J. Santoro, and R. A. Dobbins, 1993, "Aerosol Dynamic Processes of Soot Aggregates in a Laminar Ethene Diffusion Flame", Combust. Flame, 92, pp.320-333.

[45] D. L. Hofeldt, 1993, "Real-Time Soot Concentration Measurement Technique for Engine Exhaust Streams", SAE Paper No. 930079, Society of Automotive Engineers, Warrendale, PA.

[46] P. E. Bengtsson and M. Aldén, 1995, "Soot Visualization Strategies Using Laser Techniques: Laser-Induced Fluorescence in C2 from Laser-Vaporized Soot and Laser-Induced Soot Incandescence", Appl. Phys. B 60, pp.51-59.

[47] B. S. Mewes and J. M. Seitzman, 1997, "Soot Volume Fraction and Particle Size Measurements with Laser-Induced Incandescence", Appl. Optics, 36, pp.709-717.

[48] N. P. Tait and D. A. Greenhalgh, 1993, "PLIF Imaging of Fuel Fraction in Practical Devices and LII Imaging of Soot", Berichte der Bunsengesellschaft für Physikalische Chemie, 97, pp.1619-1625.

[49] S. Will, S. Schraml, and A. Leipertz, 1995, "Two-Dimensional Soot Particle Sizing by Time-Resolved Laser-Induced Incandescence", Optics Lett. 20, pp.2342-2344.

[50] D. R. Snelling, G. J. Smallwood, I. G. Campbell, J. E. Medlock and Ö. L. Gülder, 1997, "Development and Application of Laser Induced Incandescence (LII) as a Diagnostic for Soot Particulate Measurements", in press, Proceedings of the NATO/AGARD Propulsion and Energetics Panel, 90th Symposium on Advanced Non-Intrusive Instrumentation for Propulsion Engines, 20-24 October, 1997, Brussels, Belgium.

PROCEEDINGS

CSME FORUM 1998

VOLUME 1: THERMAL AND FLUIDS ENGINEERING
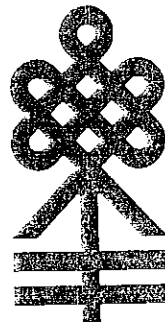
CSME/SCGM

The Canadian Society for Mechanical Engineering

La Société Canadienne de Génie Mécanique

COMTES RENDUS

FORUM SCGM 1998

VOLUME 1: GÉNIE THERMIQUE ET LE GÉNIE DES FLUIDES

RYERSON

Ryerson Polytechnic University

Toronto, Ontario, Canada

May 19 - 22, 1998

19 mai - 22 mai, 1998