Formation, Growth and Oxidation of Soot Particles
under the Influence of an Alternating Electric Field

Part II Experiments on the Sonic Sampling Technique and Premixed Flat Flames*

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Aiming at generalization of the experimental results concerning the effects of an alternating electric field on the processes of formation, growth and oxidation of soot particles reported in the previous paper, comparison between the isokinetic and sonic sampling techniques was made. The effects of an alternating electric field on the processes of soot formation were also compared between a coaxial-flow jet diffusion flame and a premixed flat flame. As a result, the superiority of the sonic sampling technique to the other technique was proved. In addition, the adverse effect of an alternating electric field on the formation of soot nuclei and its favorable effects on the coalescence, aggregation and oxidation of soot particles were reaffirmed.

Key Words: Combustion, Soot, Alternating Electric Field, Sampling Technique, Premixed Flat Flames, Coaxial-flow Jet Diffusion Flames, Electron Microscope

1. Introduction

In the previous paper⁽¹⁾ applying an alternating electric field to coaxial-flow jet diffusion flames, the size, structure and concentration of soot particles and the temperature of gas were observed at various positions within a flame. The processes of formation, growth and oxidation of soot particles were examined by comparing the results with those obtained with no electric field applied. Thus, it has been found that an alternating electric field exerts noticeable effects on both processes, and that it is possible to regulate the behavior, concentration distribution and emission of soot particles as well as the flame shape by varying the amplitude and frequency of the electric field.

However, the following problems arise when generalizations are made from the conclusions. (1) It is possible that soot particles are aggregated, disintegrated and created in the sampling probe due to the rapid cooling on sucking of burning gas into the probe.⁽²⁾ (2) Since a coaxial-flow jet diffusion flame has steep gradients of temperature, gas composition and other quantities, the mean values for different regions are obtained as the flame is oscillated by the alternating electric field, and the history of behavior of soot particles can not be traced.

An optical technique should be introduced in order to solve the first problem. However, a complete optical technique where the property of soot and the shape and size distribution of particles are fully considered

has not yet been developed. Therefore, the behavior of soot particles in the sampling process is to be inferred from the comparison between the data obtained by the isokinetic and sonic sampling techniques.

The second problem is to be dealt with by making a premixed flat flame of approximately one-dimensional structure around the axis. However, it is extremely difficult to make a flat flame stable enough even at a mixture ratio so rich that plenty of soot is produced; and if wire gauze or the like is inserted to promote the flame stability, the electric field is disturbed and the intensity is extremely reduced. Therefore, experiment has to be conducted under a condition of compromise between the mixture strength and the intensity of electric field.

In consideration of these, a trial has been made to obtain the materials for examining the data reported in the previous paper.

2. Sonic Sampling Technique

2.1 Structure of probe and sampling procedure

The structure of a probe manufactured is shown in Fig.1. The probe is made of quartz and water-cooled. The suction cone at the tip, however, is not cooled with water for prevention of soot deposit and for convenience of assemblage and disassemblage. Instead, air is ejected through the suction hole until the sampling operation to prevent it from being overheated. The suction hole is 0.2mm diam., and a stainless steel induction tube is installed to convey the soot to the filter element. This induction tube is weighed before and after the sampling operation and the amount of soot sticking on it is added to the amount collected by the filter element.

The filter sheath is made of quartz, 4.5mm i.d. and 31mm long, in which a filter element, which consists of a 0.6g/cm³ density layer of short-filament glasswool of 9μm

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Fig. 1 Sonic probe for soot sampling

diam. placed between two layers of long-filament glasswool of $10\mu\text{m}$ diam., is packed 20mm long. The deciduous filaments are removed in advance by letting air pass through the filter element for five minutes, and the filter sheath is heated in an electric furnace at 200°C for an hour, then it is weighed after cooled down gradually to room temperature in a desiccator. Since the temperature in the filter sheath rises to 90 to 160°C during sampling operation, the induction tube is weighed without drying after the sampling operation. However, the filter sheath and element are weighed after being dried in an electric furnace at 130°C for an hour and cooled down gradually to room temperature in a desiccator.

The amount of gas sampled at a time is $0.5 \sim 1$ L for diffusion flames and $2 \sim 3$ L for premixed flat flames, and the average of two runs is employed for the former. The amount of gas sampled is determined from a pressure rise in the vacuum tank (the volume is 4.4 L including the connecting tube). The observation of the size and configuration of soot particles is made using an electron microscope of penetration type (Hitachi HU-12A (125kV) or HU-11C (100kV)). For preparation of the soot particle specimens, the filter sheath and induction tube in Fig. 1 are replaced with a sheet mesh holder as shown in the figure. The sampling period is ca. 1s for a diffusion flame and ca. 5s for a premixed flat flame. The treatment of the sheet mesh and the photographic technique employed are the same as described in the previous paper⁽¹⁾

2.2 Comparison between isokinetic and sonic sampling techniques

For the coaxial-flow jet diffusion flames reported in the previous paper, comparison has been made between the distributions of soot concentrations C_s determined by two sampling techniques. The experimental condition is that the fuel used is propane; the fuel velocity $U_F = 4\text{m/s}$; the surrounding air velocity $U_A = 2\text{m/s}$; no electric field is

Fig. 2 Difference in distribution pattern of soot concentration by sampling technique (propane-air coaxial-flow jet diffusion flame with no electric field applied, $U_F = 4\text{m/s}$ and $U_A = 2\text{m/s}$)

applied; and the radial distributions have been determined at a height $h = 30\text{cm}$. The results are shown in Fig. 2. The data concerning the temperature and the isokinetic sampling technique are transcribed from Figs. 7, 8 and 9(a) in the previous paper, where the suction velocity is 4m/s .

The distribution of soot concentrations obtained by the sonic sampling technique is shifted upstream on the axis and inwards on the radius. This is probably because minute particles of yellow tarry substance which are in the initial stage of growth and easy to vaporize or oxidize are abundant on the upstream side of the temperature peak on the axis or inside the inner inflection point in the cross section, so that a considerable portion of the soot particles collected are vaporized or oxidized in the isokinetic sampling technique where the filter element reaches 300°C . In the sonic sampling technique, on the contrary, the temperature of the filter element is less than 160°C so that the reduction of soot due to oxidation or vaporization should not be significant. On the other hand, the soot particles, which have become gigantic due to growth and aggregation, can not follow precisely the rapidly contracting streamlines of a sonic sampling probe. This results in a reduction of collection efficiency in the downstream region or in the peripheral region of a flame. It is supposed, therefore, that an exact distribution of soot concentrations is close to a convolution of the distribution curves obtained by both sampling techniques.

Another thing to be considered is that it is probable in the sonic sampling technique that gas leaks in through the fit-in portion between the suction cone and the probe body due to a large pressure difference between inside and outside of the probe, and that the amount of leak increases and therefore the apparent soot concentration reduces to some extent in the downstream portion of the flame where the temperature of the suction cone falls or in the peripheral region of the flame where the probe is heated non-uniformly. If this is true, the distribution curves for both sampling techniques should come closer to each other after corrected

for leakage.

The influence of the sampling technique on the size distribution of soot particles has been examined for a propane-air coaxial-flow jet diffusion flame identical to the previous one. For convenience of the quantitative comparison of spatial transitions, comparison has been made of the volume-mean diameter $\bar{d}_v$ of spherical particles, the volume-mean diameter $\bar{D}_v$ of all particles ranging from single spherical particles to clustered agglomerates, and the mass-mean diameter $\bar{D}_w$. The results are shown in Fig.3. The experimental condition is the same as for Fig.2, and the data relating to the isokinetic sampling technique are transcribed from Figs.14 and 19(a) in the previous paper.

Fig.3 Difference in distribution pattern of mean diameter of soot particles by sampling technique (experimental conditions are the same as those for Fig.2)

By the sonic sampling technique, $\bar{d}_v$ is detected lower in the upstream region ($h < 30\text{cm}$) and higher in the downstream region ($h > 30\text{cm}$) as compared with the isokinetic sampling technique. $\bar{D}_v$ and $\bar{D}_w$, on the other hand, are detected generally lower by the sonic sampling technique except that they are detected locally higher around $h = 40\text{cm}$. The reason why $\bar{d}_v$, $\bar{D}_v$ and $\bar{D}_w$ are detected lower by the sonic sampling technique in the upstream region may be as follows. That is, since the temperature is kept rather low in the sampling probe, the evaporation and oxidation of minute particles of yellow tarry substance in the initial stage of growth are suppressed so that the possibility of their being observed is raised, and the collection efficiency is higher for smaller particles due to the contraction of streamlines. In the downstream region, on the other hand, where the fraction of huge particles increases through growth and aggregation, it is natural that the sonic sampling technique, showing a poor collection efficiency for huge particles, should give smaller $\bar{D}_v$ - and $\bar{D}_w$ -values. In addition, it is probable for the isokinetic sampling technique having a slow cooling process that the aggregation or coalescence of soot particles proceeds during the passage through the suction hole, whereas it is possible for the sonic sampling technique showing a violent shear that the clustered agglomerates of weak coupling are disrupted. The position $h = 40\text{cm}$ is in a region where the effect of suppression of evaporation and oxidation is canceled by the low collection efficiency for huge particles or vice versa. It

is natural, therefore, that $\bar{D}_v$ and $\bar{D}_w$ should have peak values around there.

In Fig.3(b) where the radial distributions of mean particle diameters are illustrated, there is only a slight difference in $\bar{d}_v$ between the two sampling techniques, whereas, for $\bar{D}_v$ and $\bar{D}_w$, the sonic sampling technique gives generally higher values. Besides, the differences are large around the peak for the isokinetic sampling technique.

Fig.4 Mean diameter distributions of soot particles on the axis with an alternating electric field applied ($E_p = 800\text{V/cm}$, $f = 400\text{Hz}$, other parameters being the same as those for Fig.2)

The axial distributions of $\bar{d}_v$, $\bar{D}_v$ and $\bar{D}_w$ are shown in Fig.4 in case of an alternating electric field applied to a coaxial-flow jet diffusion flame identical to the previous one. $\bar{d}_v$ has the same tendency as shown in Fig.3(a) in case of no electric field applied. As for $\bar{D}_v$ and $\bar{D}_w$, however, the low collection efficiency of the sonic sampling technique for huge particles is not conspicuous, since the size range is not wide enough. Reduction in size of spherical particles by the alternating electric field (see Figs.10 and 11 in the previous paper), the evaporation, oxidation, coalescence and aggregation of soot particles in the isokinetic sampling probe, and the difference in collection efficiency between particles of different sizes and disintegration of clustered agglomerates in the sonic sampling probe, all concur and present a complicated aspect.

From Eq.(6) in the previous paper, the mean diameter of spherical particles averaged over all spherical particles from single ones to ones involved in clustered agglomerates is calculated by the relation,

$$\bar{n}_p = (\bar{D}_v/\bar{d}_v)^3 \quad (1)$$

The $\bar{n}_p$ -values are evaluated from Figs.3 and 4 using the above relation and their axial transition is plotted in Fig.5. The isokinetic sampling technique gives markedly higher values, the causes of which may be as follows.

- (1) Aggregation of spherical particles proceeds in the probe.
 - (2) Minute particles of yellow tarry substance are evaporated and oxidized, vanishing from observable range.
 - (3) The collection efficiency is higher for smaller particles in the sonic sampling technique.
 - (4) Clustered agglomerates are disintegrated in the sonic sampling technique.
- As for item (2), however, both evaporation and oxidation can not be so violent as in

Fig.5 Mean number of spherical particles contained in a soot particle (experimental condition is the same as those for Figs.3 and 4)

case of concentration measurement, since the sampling period is short enough, i.e., less than 5s. In addition, the difference in n_p between the two techniques is small in the upstream region, and is even reversed locally. Therefore, it is highly probable that the progress of aggregation during the passage through the suction hole of the isokinetic sampling probe has significant effects. Especially, the peak of n_p for the isokinetic sampling technique with no electric field applied, which is located at $h = 60\text{cm}$, is difficult to explain by other than aggregation in the probe.

2.3 Discussion on the reliability of sampling technique

From the above examinations, the following conclusions may be deduced on the reliability of the isokinetic and sonic sampling techniques used for the determination of concentration or size distribution of soot particles.

As for concentration measurement, the sonic sampling technique is decisively more profitable in the region where minute particles of yellow tarry substance are dominant. However, in the region where huge soot particles such as agglomerates or clustered agglomerates are abundant, the collection efficiency falls to some extent in the sonic sampling technique where the stream tube contracts. Therefore, it would be the best to use both techniques in the right place, though the sonic sampling technique is more profitable as a whole, if precautions are taken against the leakage of air.

As for size distribution measurement, the isokinetic sampling technique is inappropriate since it is highly probable that the size distribution is significantly deformed due to the progress of aggregation during the passage through the suction hole. Although, even in the sonic sampling technique, some deformation may occur due to the low collection efficiency for huge particles and the disintegration of clustered agglomerates, far more precise results are expected than in the isokinetic sampling technique.

Although it is highly probable that the size distribution is deformed in the sampling process due to the aggregation, disintegration and preferential collection of soot particles, no evidence that soot is created in the sampling probe has been noticed.

Fig.6 Structure of premixed flat flame burner and arrangement of electrodes

3. Experiments on Premixed Flat Flames

3.1 Premixed flat flame burner

The structure of the premixed flat flame burner and the arrangement of electrodes to generate an alternating electric field are shown in Fig.6. The porous plate is a sintered brass matrix whose filtration index is $20\mu\text{m}$. The mixture feed drum of 40mm i.d. is packed with steel balls of 6mm and 11mm diam. between double 50 mesh steel gauze and a perforated plate and is cooled with water. Through the peripheral portion of the porous plate, nitrogen is to be exuded to prevent the pyrex combustion barrel of 60mm i.d. from being overheated or stained with soot. In the present experiment treating rich mixtures, however, nitrogen is turned off and a streamline controlling ring, 5mm thick and of 40mm i.d., is placed on it. Flame is stabilized between 50 mesh wire gauze placed on the ring and 50 mesh wire gauze located 25 ~ 30mm above. The probes are inserted through a hole at the center of upper wire gauze.

The electrodes for alternating electric field are a couple of aluminum boards of 100 mm x 100mm placed at 100mm interval. The nominal intensity of the field is defined as the amplitude E_p of the intensity of the electric field calculated ignoring the existence of the burner and flame. Since, however, the structure of the burner is complicated, the local intensity of the field around the axis is not known. Therefore, only qualitative discussions are possible as for the intensity of electric field.

For temperature measurement, a temperature probe is used as in the previous paper, where a Pt/Pt-13%Rh thermocouple is protruded from the quartz protection tube by 10mm and coated with SiO_2 to prevent surface reaction. Compensation for the heat loss due to radiation and conduction is omitted. The sonic

sampling technique is employed for determining the concentration and size distributions of soot particles, and the probe as shown in Fig.1 and the sampling procedure as described in Section 2.1 are used.

Pure propane for industrial purpose is used as fuel and experiments are conducted for the equivalence ratio $\phi = 1.85$ and the mixture flow rate $Q = 75\text{cm}^3/\text{s}$. It is difficult to make a flame stable enough to withstand the insertion of a probe for $\phi > 1.9$. Experiments are executed with and without an alternating electric field of $E_p = 800\text{V/cm}$ and $f = 400\text{Hz}$, and a comparison is made between the two cases.

3.2 Experimental results on premixed flat flames

A typical example of the picture of a premixed flat flame is shown in Fig.7. On the right of the picture is shown a scale indicating the distance h from the lower flame-holding gauze. Although it is not clear in the monochromatic picture, a luminous curve at the bottom corresponds to a blue flame located between $h = 2.5 \sim 3.5\text{mm}$, above that across a dark zone is seen a yellowish luminous flame between $h = 5 \sim 23\text{mm}$, and at the top comes the radiance of the upper flame-holder gauze. The diameter of the flame is ca. 50mm.

A temperature map in a plane involving the flame axis is shown in Fig.8 to visualize the one-dimensionality of the flame. Around the axis, a nearly one-dimensional flame is

Fig.7 Typical example of premixed flat flame ($\phi = 1.85$, $Q = 75\text{cm}^3/\text{s}$, no electric field applied and exposure time is $1/8\text{ s}$)

Fig.8 Temperature map in a premixed flat flame (experimental condition is the same as the one for Fig.7)

Fig.9 Axial distributions of temperatures and soot concentrations ($\phi = 1.85$ and $Q = 75\text{cm}^3/\text{s}$)

formed and temperature fluctuation is small enough.

The distributions of temperatures T and soot concentrations C_s on the axis are shown in Fig.9 with and without an alternating electric field. In both cases, the temperature has a peak around the midway between the blue flame zone and the luminous flame zone, and soot formation starts around here. This process proceeds rapidly around the luminous flame front between $h = 4 \sim 8\text{mm}$. Although the rate of soot formation is lowered a little, the concentration rises almost at a constant rate. The effect of electric field is hardly recognized, except that the temperature and soot concentration fall slightly.

The axial transition of the distribution patterns of the volume-equivalent diameters d_p and D_a , respectively, of single spherical particles and of all the soot particles is shown in Fig.10. In case of no electric field applied, particles of less than 10nm diam. are so few that the peak appears around 25nm in the size distribution. In case of electric field applied, on the other hand, the fraction of particles less than 10 nm increases so that the peak shifts to ca. 20nm. The distribution pattern is close to log-normal one except that the pattern changes gradually from Gaussian one to log-normal one in the upstream region of the flame with an electric field applied. Since it is known that soot particles, immediately after produced in the nucleation process, have a Gaussian distribution of diameters and that it shifts to log-normal one after coalescence and aggregation processes,⁽³⁾ it is noticed that the growth of soot particles due to coalescence and aggregation is suppressed by the electric field.

The volume-mean diameter $\bar{d}_v$ of spherical particles, the volume-mean diameter $\bar{D}_v$ and mass-mean diameter $\bar{D}_w$ of all the particles from single spherical particles to clustered agglomerates, and the average number n_p of spherical particles contained in a soot particle are calculated from the size distributions as shown in Fig.10 and are plotted in Fig.11. In case of no electric field applied, $\bar{d}_v$, $\bar{D}_v$, $\bar{D}_w$ and n_p are all kept at a nearly constant level, which implies that the growth and oxidation of soot particles and the formation of single spherical particles are balanced to each other. In case of an alternating electric field applied, on

the other hand, $\bar{d}_v$ is decreased and $\bar{n}_p$ is increased. The $\bar{n}_p$ -value is kept almost at one level, whereas $\bar{d}_v$ increases below $h = 15$ mm and thereafter decreases. The reason may be as follows. The amount of charge of initial compounds is changed by the electric field applied. Therefore, nucleation due to condensation of initial compounds and subsequent growth of soot particles due to coalescence and surface deposit are suppressed, so that the $\bar{d}_v$ -value at the flame front is kept down. Although the $\bar{d}_v$ -value increases thereafter through the coalescence of soot particles

caused by a forced oscillation due to the alternating electric field, the $\bar{d}_v$ -value again decreases in the downstream portion of flame since the rate of oxidation of soot particles exceeds the rate of growth. The trends of transition of $\bar{D}_v$ and $\bar{D}_w$ almost coincide with that of $\bar{d}_v$.

$\bar{n}_p$ decreases locally at $h = 5 \sim 6$ mm regardless of the application of electric field, probably because the rate of production of single spherical particles exceeds the rates of coalescence and aggregation. Especially in case of an electric field applied, $\bar{d}_v$ increases rapidly in this region, which suggests the possibility of the coalescence of minute particles of yellow tarry substance. This does not conflict with the suppression of soot growth due to the change of amount of charge, but it is probably because the frequency of collisions of single spherical particles is increased by the forced vibration by the alternating electric field. The $\bar{n}_p$ -value is considerably lower than the value for a coaxial-flow jet diffusion flames. This may be attributed to the slow progress of aggregation resulting from the soot concentration being lower by one order of magnitude than the value for a diffusion flame.

Fig.10 Axial transition of size distribution pattern of soot particles (experimental condition is the same as the one for Fig.9)

Fig.11 Axial transition of mean diameter and mean number of spherical particles (experimental condition is the same as the one for Fig.9)

3.3 Comparison with coaxial-flow jet diffusion flames

Because of various restrictions on experimental technique, only qualitative information has been obtained concerning the effects of an alternating electric field on the process of soot formation in a premixed flat flame. Since, therefore, a complete comparison with the data on coaxial-flow jet diffusion flames is difficult, only qualitative comparison and discussion are to be made below.

As for the soot concentration C_s , it is slightly decreased by the application of electric field to the premixed flat flame (see Fig.9). In the coaxial-flow jet diffusion flame, on the other hand, the peak value of C_s is markedly increased both on the axis and in the corresponding cross section by the application of an electric field (see Figs.7 to 9 in the previous paper). In particular, the increase of the soot concentration integrated over the corresponding cross section is prominent. This fact implies that the effects of an alternating

electric field on the processes of soot formation and growth are completely different between the two flames. This is probably because the direction of vibration of soot particles by the electric field is parallel to the isothermal lines or isoconcentration lines in a flat flame and perpendicular in diffusion flames. Although a diffusion flame fluctuating under the influence of an alternating electric field shows gently-sloping distributions of temperatures and soot concentrations if observed in time-averaged manner, it should have steep gradients around the flame surface instantaneously. According to the investigation of Kono et al.⁽⁴⁾ who applied an alternating electric field normally to a

premixed flat flame, the flame luminosity increases with the intensity of the field if the amplitude of oscillation of soot particles is within a certain limit, which implies an increase in concentration of soot. If there is any difference between a diffusion flame and a premixed flame, it is highly probable that the effects of an alternating electric field on the process of soot formation and growth depend on the direction of the field.

As for the size distribution, an electric field suppresses soot nucleation by the condensation of initial compounds and subsequent growth of soot particles by coalescence and surface deposit in a flat flame, so that the diameter d_v of spherical particles is reduced as noticed from Figs.10 and 11. Although the coalescence and aggregation of soot particles also occur in terms of the forced oscillation of soot particles by the alternating electric field, d_v starts to decrease again before it increases enough due to the accelerated oxidation. Since, however, the frequency of collisions increases due to forced oscillation, coalescence of spherical particles proceeds and n_p increases to some extent. Concerning diffusion flames, on the other hand, Figs.12 and 13 in the previous paper exaggeratedly correspond to Fig.10 in the present paper, whereas Figs.14 to 16 in the previous paper or Figs.3 to 5 in the present paper show essentially the same tendency as Fig.11 for a flat flame. In addition, it is Figs.10 and 11 in the previous paper that visualize directly the effects of an alternating electric field on the configuration and size of soot particles.

4. Conclusions

Aiming at generalization of the experimental results about the effects of an alternating electric field on the processes of formation, growth and oxidation of soot particles reported in the previous paper, comparison between the isokinetic and sonic sampling techniques has been made. In addition, the variation of the processes of soot formation and growth has been observed by applying an alternating electric field to a premixed flat flame. The principal results are as follows.

(1) As for soot concentration measurement, the sonic sampling technique gives more precise information in a region abundant in minute particles of yellow tarry substance. In a region abundant in huge particles, on the other hand, the isokinetic sampling technique is more profitable, since the collection efficiency decreases there in the sonic sampling technique. As a whole, the sonic sampling technique is superior, if precautions are taken against the leakage of air.

(2) For the purpose of the determination of size distribution of soot, the isokinetic sampling technique, in which, due to the slow cooling during the passage through the suction hole, the evaporation and oxidation of minute particles of yellow tarry substance and the growth of soot by the aggregation of soot particles take place, is inadequate. Since, even in the sonic sampling technique,

the collection efficiency varies with particle diameter, some errors may occur, but far more precise size distribution data than from the isokinetic sampling technique are expected from the sonic sampling technique.

(3) No evidence that soot is created in the probe has been noticed.

(4) The effects of an alternating electric field on the soot concentration are different between a coaxial-flow jet diffusion flame and a premixed flat flame, that is, the soot concentration is increased in the former and decreased in the latter. This is probably because the alternating electric field acts at right angles in the former, and in parallel in the latter, to the isothermal lines or isoconcentration lines around the flame front.

(5) The effects of the alternating electric field on the size distribution of soot particles are essentially the same between a diffusion flame and a flat flame. That is, an alternating electric field suppresses soot nucleation by the condensation of initial compounds and subsequent growth of soot particles by coalescence and surface deposit, but at the same time, it accelerates the coalescence and aggregation by making soot particles oscillate forcibly and form agglomerates of complicated structure and clustered agglomerates. Occasionally the field accelerates the oxidation resulting in a reduction in diameter of spherical particles.

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