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SOOT PRODUCTION BY DIFFUSION FLAMES

by

I. S. McLINTOCK

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Fire Research Station.
Boreham Wood.
Herts.
(phone ELStree 1341)

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Part 1. Summary of Available Information and
Discussion of its Relevance
to Fire Problems

SECTION 1.

Introduction

The production of smoke in fires constitutes a serious hazard. Smoke can drastically reduce visibility and has unpleasant physiological effects. In addition, the heat radiated by flames is related to the concentration of radiating solid particles within the flame. It would be useful to have some information on those factors which might influence smoke production by flames.

A major component of smoke from flames is soot, a solid material generally of high carbon content. This material makes a large contribution towards its obscuring power. Information on the rate of soot production by diffusion flames under various combustion conditions would therefore be appropriate to an investigation of the smoke production problem. At present there is surprisingly little quantitative information on the rate of soot production, even under laboratory conditions.

In enclosed compartments the composition of the atmosphere in which flaming combustion is taking place will not, in general, correspond to that of air. The composition will be modified by the presence of combustion products and decomposition products; the use of inerted atmospheres or of chemical extinguishants to control the fire will cause further modification of the atmospheric composition. Smoke production might be expected to vary with such changes in combustion conditions and it would therefore be useful to investigate rates of smoke production when different diluents are added to a flame. Measurement of sooting rates, rather than smoking rates, would probably be experimentally simpler.

The literature on soot formation in flames is extensive and the results are often conflicting. In part this is due to the variety of systems which have been investigated. Reference to Appendix 2 and Section 3 of this report will show that the conflict is particularly pronounced as far as the effects of additives and diluents are concerned. Most investigations have been concerned with measurements of "smoke points", i.e. the critical fuel flow (diffusion flames) or mixture strength (premixed flames) at which soot just begins to leave the flame. Quantitative information is confined, for the most part, to measurements of carbon inside the flame. In the present context we are concerned, rather, with the quantities of unburnt carbon which leave the flame.

It should be made clear at this point that soot produced by flames is not elementary carbon. It may contain hydrogen and oxygen in atomic percentages in excess of 30%, particularly under mild combustion conditions⁽¹⁾⁻⁽⁵⁾. Briefly, soot produced under mild combustion conditions has the characteristics of a polybenzenoid hydrocarbon; it may contain volatiles of lower molecular weight,

some of which may possess oxygenated functional groups. Under stronger combustion conditions a soot of higher carbon content, and with interstitial hydrogen and oxygen set in a strained graphite lattice, is formed. In this report the term "soot" is used to cover solid material given off by flames. The word "carbon" will be used to describe such material inside the flame, on the understanding that such a term may not adequately describe its chemical nature.

It will be appropriate to begin with a brief mention of current ideas on the mechanism of soot formation in flames. Relevant information on those factors which are known to influence soot production will then be summarised. Both diffusion and premixed flames will be discussed.

SECTION 2.

Mechanism of soot formation in flames

Many theories of carbon formation have been proposed. Some have later been discarded as untenable but even at present the problem has by no means been resolved. References to work in this field are presented in Appendix 1, as are references to the many adequate reviews of the subject.

It is generally agreed that the process of carbon formation in flames is extremely rapid; simple fuel molecules are converted to aggregates of several thousand carbon atoms in times of the order of milliseconds. Such a process is almost certain to involve free radicals. Electron spin resonance measurements on soot from diffusion flames have, in fact, demonstrated the presence of unpaired electrons in the soot⁽¹⁾⁽³⁾. This may be taken as circumstantial evidence for a free radical mechanism.

The process of carbon formation is now considered⁽¹⁶⁾⁽²³⁾ to involve two stages:

- (i) Conversion of fuel to carbon nuclei.
- (ii) Growth of carbon nuclei by a heterogeneous process.

A third process:

- (iii) Oxidation of carbon particles in a heterogeneous reaction with oxidants such as CO_2 , H_2O , O_2 and OH , competes with these two carbon formation reactions.

In this context diffusion and premixed flames differ in an important respect. In a premixed flame the oxidation processes (carbon removal) proceed concurrently with the carbon formation processes; in a diffusion flame carbon formation is generally well established before the competing reactions become significant. Hence a diffusion flame can exhibit luminosity due to hot carbon particles within it, but yet produce no soot. On the other hand, in premixed flames the onset of luminosity and soot production frequently coincide.

Whether a flame soots or not depends on the balance⁽²²⁾⁽²⁷⁾⁽³¹⁾ between these two competing processes. A diffusion flame will give off soot (or a premixed flame will show carbon luminosity) if the overall rate of carbon formation exceeds that of carbon combustion.

Investigations have been carried out on the separate processes of carbon formation by hydrocarbon pyrolysis⁽⁶⁾⁻⁽¹⁶⁾ and of carbon particle combustion⁽¹⁷⁾⁻⁽²¹⁾. Even under these circumstances the results are not straightforward. In a flame the situation will be further complicated by such factors as flame size and structure and the considerable temperature gradients which exist in the flame.

SECTION 3.

Effect of additives

Published results on the effect of additives on soot formation show a large

measure of disagreement. It has been mentioned earlier that most work has been concerned with the effect of additives on smoke point⁽²⁶⁾⁽²⁷⁾ and luminosity⁽⁴⁾⁽¹⁴⁾⁽²⁴⁾⁽²⁵⁾⁽³⁵⁾. Milberg⁽²²⁾ has measured sooting rates of acetylene-air diffusion flames at fuel flows beyond the smoke point, but not the effect of additives on these rates. The effect of additives on the rate of carbon formation within non-smoking diffusion flames has been measured⁽²³⁾ and similar experiments have been carried out with premixed flames⁽²⁷⁾.

In the case of diffusion flames additives have generally been introduced to the fuel stream rather than to the air stream. The results of Creitz⁽³⁶⁾ are of interest here, although they refer to the extinction of diffusion flames by halogenated methanes; the effectiveness of the extinguishant depended on whether it was introduced to the fuel or the air stream. When large amounts of additive are employed it is frequently termed a "diluent", especially if the oxygen concentration is thereby depressed significantly.

The complexity of, and conflict between, the available results is summarised in Appendix 2.

The mechanism by which "inert" additives such as nitrogen affect soot formation has not yet been adequately explained. One obvious effect of introducing such an additive will be a lowering⁽⁴⁾⁽²⁴⁾ of flame temperature. This change in flame temperature has generally been discussed⁽⁴⁾⁽²⁴⁾ in relation to the carbon formation reaction rather than to the overall soot production process. Such an approach would not appear to be adequate, since the decrease in flame temperature will lower the rates of both carbon formation and combustion. In theory it would be possible to calculate which rate was decreased by the larger amount and hence the effect (on sooting rate) of a known decrease in flame temperature. The situation is complicated in practice by at least two factors:

(i) Information is available on the activation energies of carbon formation by hydrocarbon pyrolysis⁽⁶⁾⁽⁸⁾⁽¹⁶⁾⁽²⁹⁾⁻⁽³¹⁾ and of carbon combustion⁽¹⁷⁾⁽¹⁹⁾⁽²⁰⁾⁽³⁷⁾. But Tesner⁽²⁰⁾ has shown that the activation energies of both types of reaction increase with decrease in temperature. The effect of temperature on rate will therefore be complex.

(ii) The presence of temperature gradients throughout the flame will aggravate the difficulties referred to above.

The calculation appears to be intractable at present. The effect of flame temperature on carbon and soot formation has been examined experimentally. The results are summarised in Section 4.

In any case, however, several results⁽⁴⁾⁽²⁴⁾ indicate that the effect of "inert" diluents on carbon or soot formation cannot be explained purely on the basis of their addition resulting in a decrease in flame temperature. Street and Thomas⁽²⁴⁾ have discussed the situation in terms of a decrease in oxygen concentration. Arthur has suggested⁽³²⁾ that hydrogen atoms play an important role in carbon formation and that additive molecules suppress carbon formation by acting⁽³²⁾ as third bodies in hydrogen atom recombinations. Later work⁽⁴⁾ by Arthur and Napier shows that, although hydrogen atoms may be important species in the reactions which lead to carbon formation, the situation cannot be adequately explained on this basis.

It will be useful here to speculate briefly on further possible explanations of the effect of inert additives. It has been mentioned that carbon formation in flames is almost certain to involve a free radical mechanism. The free radicals involved are likely to be those encountered in typical hydrocarbon pyrolyses as well as large polybenzenoid precursors of soot particles. It seems reasonable that in such a system inert molecules such as nitrogen may alter the radical concentrations by acting as second or third bodies in collisional processes. In this role they would play a direct physico-chemical part in the carbon formation process.

Some information is available⁽⁵⁾⁽³³⁾⁽³⁴⁾ on the concentrations of nitrogen and combustion products (CO_2 , H_2O , CO) within diffusion flames. Nitrogen concentrations inside a flame may be as high as 80%, even when it has not been added as a diluent. It is therefore suggested that nitrogen molecules may influence the rate of carbon formation in all diffusion flames burning in air. Nitrogen introduced as a diluent could reinforce this effect. Molecules of combustion products might be expected to behave similarly, but they might influence carbon formation by additional, more obviously chemical, effects.

Other additives mentioned in Appendix 2 cannot be classed as inert. Various chemical explanations of their effect on soot formation have been suggested. For example, the promoting effect of SO_3 could be a result of increased radical formation following the formation of peroxides between fuel molecules and SO_3 ⁽³⁵⁾. It is considered⁽³⁵⁾ that SO_2 may suppress soot formation by reacting with solid carbon as well as by reacting with free radicals involved in the carbon formation process. Other additives may be considered to operate as sources of free radicals; appropriate references to halogens and halogenated compounds are given in Appendix 2. Additives such as CO_2 , H_2O and CO may alter flame chemistry by upsetting equilibrium reactions in the flame, and CO_2 is involved further in the carbon combustion reactions.

The introduction of large amounts of diluent will decrease the oxygen concentration in the flame. This might be expected⁽³⁷⁾ to lower the rate of carbon combustion. Small amounts of oxygen are known to accelerate⁽²⁸⁾ the rate of hydrocarbon pyrolysis. Pyrolysis of hydrocarbons is involved in several of the current mechanisms of carbon formation and it is therefore possible that changes in oxygen concentration will affect the rate of carbon formation as well as that of carbon combustion.

SECTION 4.

Effect of some other variables

(a) Air flow. Smoke points of diffusion flames increase with increase in air flow (for a given burner diameter) up to a critical air flow beyond which increases in air flow have no further effect in suppressing smoke formation⁽²⁶⁾. The value of this critical air flow is specific for a given system. Milberg has shown⁽²²⁾ that rates of soot production for acetylene-air diffusion flames decrease with increase in air-fuel ratio.

(b) Temperature. Schalla and McDonald report⁽²⁶⁾ that increase in the flame temperature of diffusion flames either increased smoke points monotonously or else lead to an initial decrease in smoke points followed by a steady increase. The fuel used determined which of the two effects was observed. Their method of altering flame temperature, however, was to add varying amounts of nitrogen or argon to the oxidant flow. In the light of the discussion in Section 3 above it is possible that they thereby altered other parameters as well as flame temperature.

According to Street and Thomas⁽²⁴⁾ increase in the flame temperature of premixed flames decreased their luminosity and therefore decreased carbon formation.

(c) Pressure. Soot production increases with pressure⁽²²⁾⁽²⁶⁾⁽²⁷⁾ for both diffusion and premixed flames. In any particular case there is a critical pressure below which soot formation does not occur.

SECTION 5.

Conclusions

There are several gaps in our knowledge of the processes of soot formation in diffusion flames. In particular, information on the variation in sooting rate with combustion conditions is very meagre. Further information of this

kind, with particular reference to the part played by atmospheric composition, should prove useful in a study of the problem of smoke production by fires. A systematic study, on a laboratory scale, of the effect of fuel type, diluents and temperature on soot production in flames would be appropriate.

To complete the investigation it would be necessary to study the effect of these factors on soot, or smoke, production during smouldering combustion.

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APPENDIX I

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Notes

Isotopic tracer studies; application
to C_2H_2 mechanism and to $2CO \rightleftharpoons C_s + CO_2$
mechanism.

Rate of soot growth in turbulent flow
with combustion products and methane.

APPENDIX II

Effect of additives on carbon and soot formation

Additive or diluent	System	Effect	Reference
N ₂	Diffusion flames	Suppression	(a),(b),(d),(e)
	Diffusion flames; carbon within flame	Suppression (but dispersion increased)	(c)
	Hydrocarbon pyrolysis	Suppression	(f)
	Diffusion flames	Promotion	(t)
	Premixed flames	Promotion <u>or</u> nil	(g),(h)
A and He	Premixed flames	Suppression	(g)
H ₂	Diffusion flames; carbon within flame	Suppression (but dispersion increased)	(c)
	Hydrocarbon pyrolysis	Suppression	(f)
	Premixed flames	Suppression	(g),(h)
	Premixed flames (addition of H atoms)	Promotion	(i)
	Diffusion flames	Suppression	(d)
CO ₂	Diffusion flames	Suppression	(a),(b),(e),(d)
	Premixed flames	Suppression	(g)
	Premixed flames	Nil	(h)
CO	Diffusion flames	Suppression	(b),(d)
	Premixed flames	Promotion	(j)
	Premixed flames	Suppression	(j),(h)
	Premixed flames	Nil	(g)
SO ₂	Diffusion flames	Nil (suppression in very rich flames)	(h)
	Diffusion flames	Suppression	(b),(s)
	Premixed flames	Suppression	(k)
SO ₃	Premixed flames	Promotion	(g),(k),(h)
	Diffusion flames	Suppression?	(j)
CS ₂	Premixed flames	Nil (or slight promotion)	(h)
H ₂ S	Premixed flames	Suppression	(k)

APPENDIX II (Contd)

Additive or diluent	System	Effect	Reference
H ₂ O	Premixed flames	Suppression	(g)
	Premixed flames	Nil	(h)
	Diffusion flames	Suppression	(d)
Halogens and halogenated compounds	Premixed flames	Promotion	(h)
	Premixed flames (addition of halogen atoms)	Promotion (also for other atoms and radicals)	(i)
	Diffusion flames	Promotion	(l),(n),(s)
	Premixed flames	Very small effect	(k)
Argon	Diffusion flames. See reference	Results complex	(m)
NO, N ₂ O NO ₂	Diffusion flames	Promotion or nil	(p)
	Diffusion flames (coal).	Suppression	(q)
Others			(c),(g),(h), (q),(r)

Reference to Appendix II

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