Extinction Properties of Smoke Mixtures

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ABSTRACT

A convenient and basic measurement on smoke produced by a given material is the specific extinction area. It is, in theory, the more appropriate quantity by which to attempt correlation between small and large-scale data. In fact, it is now well established that the smoke yield and the soot particle size are increased as the fire size increases. In general, procedures on how to integrate such complex phenomenon, as particle surface grow process as well as coagulation and coalescence, into correlation, have not yet been proven. Despite this difficulty, the present authors began a basic small-scale study on extinction properties of mixtures of smokes, using the concept of specific extinction area. A theoretical relationship between the specific extinction areas of the different fuels involved (burning side by side, or one in the flame of another) has been established. Dynamic measurements with a flow-through system have been made. Comparison with the theoretical development shows a satisfying agreement.

INTRODUCTION

There is an extensive literature dealing with smoke production and properties, and general information on this problem may be gleaned in many detailed reviews or textbooks (for instance [1] and [2]). Most investigations have been concerned with the measurement of smoke production potential and burning tendency of materials, and especially with smoke optical properties. This latter aspect is important as far as fire safety is concerned: by reducing visibility, smoke can present great threat to persons. But optical properties measurement is also important when studying radiation from smoke that can largely affect heat flux distribution and heat balance of a fire.

Production and optical properties of smoke are the most often measured simultaneously. The most conventional and suitable method for optical measurement purpose consists in measuring the attenuation of monochromatic light beams over a given path length of smoke. However, the deduced extinction coefficient ke_λ may depend not just on what material is being burned but also on the burning characteristics and environment. It is an extensive property which can be expressed as the product of the mass concentration of the smoke particulate C (where the attenuation is being measured) and of an extinction coefficient per unit mass concentration $\sigma_{s\lambda}$ referred to as a specific extinction area on smoke particle-mass basis:

$$ke_\lambda = C\,\sigma_{s\lambda} \tag{1}$$

In fact, to estimate smoke emission from a fire, a parameter of more practical relevance is the specific extinction area on fuel mass-loss basis $\sigma_{f\lambda}$, the quantities $\sigma_{s\lambda}$ and $\sigma_{f\lambda}$ being related through:

$$\sigma_{f\lambda} = \varepsilon\,\sigma_{s\lambda} \tag{2}$$

where ε represents the mass of smoke particles generated per mass of fuel consumed.

Therefore, in a dynamic system, the specific extinction area $\sigma_{f\lambda}$ can be defined by:

$$\sigma_{f\lambda} = \frac{ke_\lambda\,\dot{q}}{\dot{m}} \tag{3}$$

where $\dot{m}$ is the fuel mass-loss rate and $\dot{q}$ is the smoke volume flow rate.

Independent of test conditions (changes in burning rate and dilution), the quantity $\sigma_{f\lambda}$ is a basic measurement on smoke that could be expected as a proper variable by which to attempt correlation between small- and large-scale data. However, the fact that the specific extinction area $\sigma_{f\lambda}$ is a constant for a given fuel is a theory that is being disputed by investigators. Extensive reviews found any or few correlation between small- and full-scale data (for instance [3] to [5]. A noticeable increase in smoke yield with increasing fire size is generally observed. Based on results from experiments on crude oil pool fires from 0.085 to 17.2 m in diameter, Notarianni et al. [6] show that smoke yield varies approximately by a factor two between these small and large-scale tests. Recent experiments conducted by Mulholland et al. [7] on the properties of smoke produced likewise by crude oil fires confirm the trend of increasing smoke yield by about 50% with increasing pan size.

Only some authors have shown promising trends. It has been shown that smoke obscuration in full-scale fires is a function of both the specific extinction area and the amount of fuel burnt which can be characterised in terms of heat released. Bard and Pagni [8] have proposed a correlation in regard to the maximum smoke emission. For solid and liquid fuels, it was found by Mulholland et al. [9] that large-scale emission rate could be well represented from cone calorimeter data of the material mass-loss rates per unit area matched for the two cases. However, even if the prediction of full-scale smoke obscuration from small-scale data is not yet addressed properly, a research effort has to be carried on to describe smoke release and obscuration in most of the current fire situations.

More often, only a single fire source is taken into consideration. But in reality, in many fire situations, several fuel areas are involved in burning and smoke production. As a fire grows, flames can spread to nearby combustible items and surfaces, smoke being consequently generated by different materials. Moreover, a material may burn engulfed within the flame from another material. Unfortunately, information concerning this problem is scarce and there is so far no predictive analysis or relationship relative to mixtures of smokes. The present work was undertaken with the following objectives:

- develop a general functional relationship relative to smoke mixtures on the basis of smoke area concept, which remains the more convenient parameter for smoke measurement.
- measure this basic quantity with a well-suited open flow-through procedure, for smoke generated by various types of liquid and solid fuels, and validate such a relationship.

THEORETICAL CONSIDERATIONS RELATED TO SMOKE MIXTURES

Knowing that the average extinction coefficient ke_λ can be expressed in terms of particle number density N, effective cross section s, and monochromatic extinction efficiency Qe_λ as:

$$ke_\lambda = N\,s\,Qe_\lambda \tag{4}$$

it can be further written:

$$ke_\lambda = \frac{C}{\rho\,v}\,s\,Qe_\lambda \tag{5}$$

where C is the mass concentration, ρ the density and v the volume of smoke particles (assumed to be spherical). However, the mass concentration of smoke particles can also be expressed as:

$$C = \frac{\varepsilon\,\dot{m}}{\dot{q}} \tag{6}$$

Thus, introducing the diameter of smoke particles d, it comes for the extinction coefficient:

$$ke_\lambda = \frac{3\,\varepsilon\,Qe_\lambda\,\dot{m}}{2\,\rho\,d\,\dot{q}} \tag{7}$$

and, substituting this expression of ke_λ in equation (3), the specific extinction area $\sigma_{f\lambda}$ is expressed as:

$$\sigma_{f\lambda} = \frac{3\,\varepsilon\,Qe_\lambda}{2\,\rho\,d} \tag{8}$$

It has experimentally been shown that for a wide range of combustible materials the group $\frac{3\,Qe_\lambda}{2\,\rho\,d}$ is nearly constant (the particles generated by the different fuels are quite similar in density and size).

Therefore, $\sigma_{f\lambda}$ is expected to be a constant for the smoke produced by a given material [10].
Let us consider now a mixture of smokes generated by different fuels, each of them being characterised by a given yield of particles ε_i and a given mass-loss rate $\dot{m}_i$. The smoke particle mass concentration becomes:

$$C = \frac{\sum_i \varepsilon_i \dot{m}_i}{\dot{q}_m} \qquad (9)$$

($\dot{q}_m$ being the volume flow rate of the smoke mixture). The specific extinction area of the mixture is expressed as:

$$\sigma_{fm_\lambda} = \frac{\sum_i \varepsilon_i \dot{m}_i}{\sum_i \dot{m}_i} \frac{3Qe_\lambda}{2\rho d} \qquad (10)$$

and the specific extinction area σ_{fi_λ}, characterising the different smokes, can then be related by:

$$\sigma_{fm_\lambda} = \frac{\sum_i \dot{m}_i \sigma_{fi_\lambda}}{\sum_i \dot{m}_i} \qquad (11)$$

This expression gives a proper weight-averaged specific extinction area. Some experimental validations are proposed further on. It is noteworthy that for fire smokes the wavelength effect appears marked. The simple empirical relation found experimentally, $\sigma_{f\lambda} = A\,\lambda^{-n}$ (where A is a constant and n and exponent generally close to one), reveals small size, mainly absorbing particles.

EXPERIMENT

Chosen Fuels

Heating oil has been the selected liquid fuel because it is known to have a high sooting propensity and it is frequently used as a reference in laboratory studies. The solid materials were chosen according to their fire performance and to their ability to generate a relatively less production of smoke than the considered liquid fuel. Polymethylmethacrylate (PMMA), for instance, does not melt or drip significantly while burning and is therefore convenient to handle. On the other hand, since it is an oxygenated fuel, it gives substantially less smoke than hydrocarbon liquids.

In other respects, new trends in construction and the growing need in electric equipment lead to use a large number of materials treated to become fire-retarded. Generally, these materials cannot self-sustain a flame but can burn if supported by intense heat flux from another source (the flame of a nearby fire or a radiant external surface). It is the case, for instance, of most electric cables used in high-risk areas where a fire accident must be avoided, and especially in nuclear power generating plants. Then, two types of fire-retarded electric cables (EPR and PVC cables), most commonly used in the "Electricité de France" production units are tested. EPR stands for ethylene-propylene rubber copolymer (Hypalon is a trademark). The main polymeric material of the sheathing is chlorosulphonated polyethylene. It is laden with mineral fire-retardants. The insulation is based on reticulated ethylene-propylene coating three conductor wires made of copper, whose nominal cross-sectional area is 1 mm^2. PVC sheathing and insulation (resin and plasticiser) are laden with antimony oxide and alumina trihydrate. The insulation coats four conductor wires of copper, whose nominal cross-sectional area is 1.5 mm^2.

Experimental set-up for the determination of the specific extinction areas

The small-scale experimental set-up is an original flow-through system allowing an accurate control of the above-mentioned three parameters (fuel mass-loss rate, smoke extinction coefficient and volume flow rate). The main features of the procedure have previously been described [11]. The smoke measurements (light attenuation and volume flow rate) are made near the exit of the exhaust duct of the flow-through system, in the stream section where a well calibrated jet of smoke is generated. The velocity is measured with a Pitot probe and the extinction coefficient is classically measured through the attenuation of horizontal light beams for three wavelengths of the visible spectrum: 488 nm (argon laser), 633 nm (helium/neon laser, 780 nm (laser diode). The path length coincides with the section diameter of the exhaust duct (9.8 cm). Each beam is received on a photodiode detector through a narrow-band filter to eliminate external noise.

Equipment for fuel mass-loss measurement depends on whether the fuel is a liquid or a solid. Liquid fuels are contained in circular 5 cm deep steel pans of different diameters (8 or 10 cm). Notice that these diameters are in the transitional regime, where it is well known that the burning rate increases with the size of the pan [12]. Thus, two values of mass-loss rate can be experienced. The determination of this quantity is made by means of a constant level gravity liquid feeding system based on an electronically controlled on/off valve [13]. After a transient period, a steady burning is rapidly reached and a nearly constant value of the specific extinction area (Eq. 3) can be obtained.

Slightly more complex is the burning of a solid fuel. There is no equivalence to the simple evaporation process of the liquid. Flaming combustion involves irreversible and, more often unsteady chemical decomposition, and the burning behaviour is modified accordingly. In addition, burning rate exhibits generally three stages: a growth stage (the flame spreads out progressively on the entire material surface), a fully developed stage where a maximum is reached, and a decay stage. During the growth and decay stages, the smoke particle concentration in the flow can be non homogeneous. It therefore appears more convenient to characterise such materials by integrating data over the total period of smoke production. In such conditions, the total specific extinction area is obtained by:

$$\sigma_{f\lambda} = \frac{1}{\Delta t} \int_{\Delta t} \frac{k_{e\lambda}\, \dot{q}}{\dot{m}_s} dt \qquad (12)$$

the instantaneous mass-loss rate $\dot{m}_s$ being recorded with a load cell on which samples are placed.

PMMA samples are sticks, 12 cm in length and 2.5 cm in diameter, accommodated by a sample holder to burn in horizontal position (Figure 1(a)). Electric cables (EPR and PVC) samples are sticks, 12 cm in length and 1.85 and 1.50 cm in diameter respectively, accommodated by a sample holder to burn in two orientations: horizontal and vertical (Figure 1 (a) and (b)). A small butane line burner provides the flame assistance for their burning.

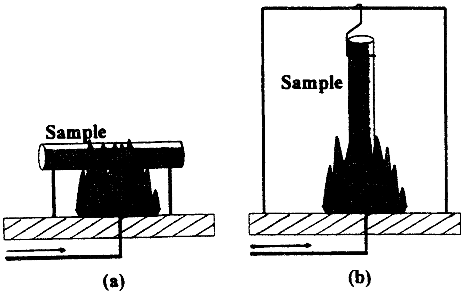


FIGURE 1. Solid sample holder and flame assistance: (a) horizontal orientation, (b) vertical orientation.

The experimental set-up can be used either with a single fire source: one liquid, one solid, or a solid burning in the flame of a liquid (Figure 2 (a)), or with two fire sources: two liquids, two solids, or a liquid and a solid, located side by side (Figure 2 (b)).

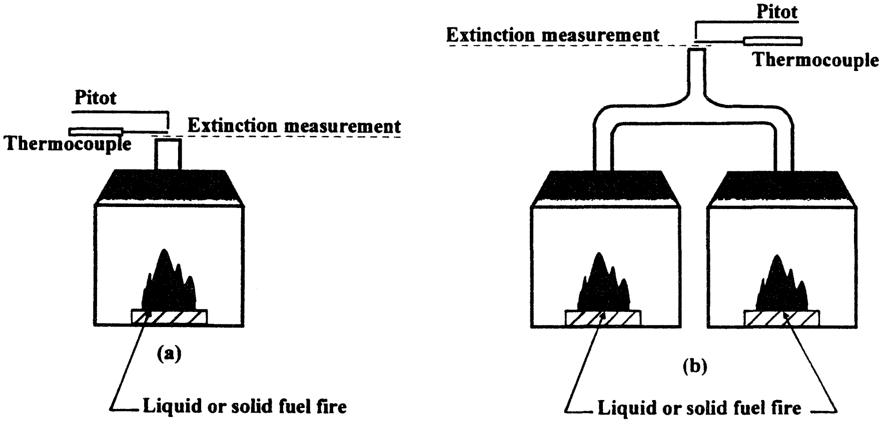


FIGURE 2. Experimental device: (a) single fire source, (b) two-fire source.

If smoke is produced by two liquid fires, (two pans of heating oil of different size), once steady burning conditions are reached, the specific extinction area of the mixture σ_{fm_λ} is determined by:

$$\sigma_{fm_\lambda} = \frac{ke_{m\lambda}\,\dot{q}_m}{\dot{m}_{l1} + \dot{m}_{l2}} \qquad (13)$$

$\dot{m}_{l1}$ and $\dot{m}_{l2}$ being the two fuel mass-loss rates.

If smoke is produced by a liquid fire and a solid fire (a pan of heating oil and a stick of PMMA for instance), a summation according to (12) must be made:

$$\sigma_{fm\lambda} = \frac{1}{\Delta t} \int_{\Delta t} \frac{ke_{m\lambda}\, \dot{q}_m}{\dot{m}_l + \dot{m}_s} dt \qquad (14)$$

$\dot{m}_l$ and $\dot{m}_s$ being respectively the instantaneous mass-loss rates of the liquid and solid fuels.

RESULTS AND DISCUSSION

Distinct fires

- **Smoke generated by two liquid fires**

The three parameters required to calculate the specific extinction area have been measured first for two heating oil pool fires (8 and 10 cm in diameter), and next for the smoke mixture. Data are reported in Table 1. They appear to fit reasonably well the theoretical relation (11). This shows obviously, as stated above, that, if there is no change in the properties of particles and in the yield of smoke, the specific extinction area is conserved.

TABLE 1. Summary of the results obtained with two pans of heating oil of different size burning side by side.

Specific extinction area	Heating oil 8 cm pan	Heating oil 10 cm pan	Mixture	Relation (11)
σ_{488} (m^2.kg^{-1})	1497	1464	1415	1475
σ_{633} (m^2.kg^{-1})	1032	997	1135	1009
σ_{780} (m^2.kg^{-1})	877	848	899	858

These tests show the ability of such a small-scale procedure, with simple flow geometry, to predict the extinction properties of smoke mixtures. Insofar as the burning rates are different for the two pans, while the group $\frac{3\,Qe_\lambda}{2\,\rho\, d}$ can be considered to be a nearly constant property of the smoke particles, the dependence of the particle concentration on air entrainment, i.e. dilution, is evidenced.

- **Smoke generated by a liquid and a solid fire**

The specific extinction area has been determined as above, at first for a stick of PMMA and next for the mixture of smoke generated by the 10 cm pan of heating oil and the stick of PMMA. Data are reported in Table 2. A satisfying agreement with the theory is likewise observed.

TABLE 2. Summary of the results obtained with a pan of heating oil and a stick of PMMA burning side by side.

Specific extinction area	Heating oil 10 cm pan	Stick of PMMA	Mixture	Relation (11)
σ_{488} ($m^2.kg^{-1}$)	1464	270	803	778
σ_{633} ($m^2.kg^{-1}$)	997	189	510	493
σ_{780} ($m^2.kg^{-1}$)	848	156	408	418

It is noteworthy that the value of the resulting specific extinction area is appreciably less than the one obtained for the fuel having the highest sooting propensity. This shows clearly that, in addition to simple dilution effects, balanced compositions are achieved by mass-loss rates.

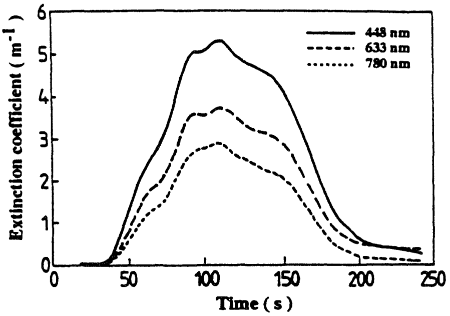
Solid burning in the flame of a liquid fuel

The study of smoke mixtures generated by solids burning engulfed in the flame of a liquid pool fire, is made by combining heating oil pool fire and sample of electric cables. In this case, the experimental procedure consists in two steps: burning of heating oil alone until a steady state regime is reached, and next, insertion of the sample of solid into the flame. The insertion of the sample into the flame results in an increase of the extinction coefficient and volume flow rate, showing an additional production of smoke. Tests have been made using the 10 cm diameter heating oil pool fire and sticks of EPR cables positioned horizontally or vertically, as explained above.

A comparison with the values obtained by means of the theoretical relation (11) can be made using the specific extinction area relative to the heating oil burning in the 10 cm pan (see Table 1 or 2), and to the sample of EPR cable. This comparison is reported in Table 5, coming after some considerations relative to the combustion of EPR cable alone.

As aforementioned, the cables are laden with fire-retardants and cannot burn without an external calorific assistance. Thus, the specific extinction area of this cable has been determined using the assistance of a small line butane burner (see Fig. 1). The power of the burner must be accurately chosen. If it is too low, the burning rate of the cable is not strong enough to obtain a homogeneous smoke composition in the free stream section near the exhaust duct of the experimental device (see Fig. 2 (a)) and, therefore, to have reliable measurements of attenuation. On the other hand, for sample burning in vertical orientation, if the heat flux of the burner is too high, the external sheath can be distorted, teared and splitted so that some part of this sheath can be outside the pilot flame and consumed in non-flaming combustion (smouldering), giving rise to smoke particles whose characteristics are quite different from those produced by flaming combustion. A less intense thermal assistance results logically in a less regular combustion but in a greater time of combustion and, in fact, leads to more reproducible measurements.

Figure 3 shows a typical evolution of the extinction coefficient with time for a sample of EPR cable burning in horizontal orientation, while Figure 4 shows the same evolution for a sample burning in vertical orientation. In the latter case, the phenomenon of distortion and splitting of the external sheath with renewal combustion is clearly evidenced.



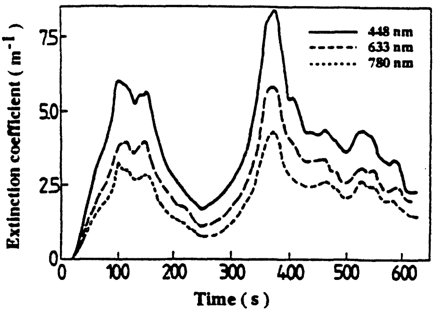


FIGURE 3. Evolution of the extinction coefficient as a function of time for a sample of EPR cable burning in horizontal orientation.

FIGURE 4. Evolution of the extinction coefficient as a function of time for a sample of EPR cable burning in vertical orientation.

The recordings show different steps of combustion: burning of the external sheath, tearing and widening of this one at the bottom, giving rise to a revival of combustion due to an increase of the burning surface. As a consequence, the values of the specific extinction area obtained in vertical orientation are, more often, slightly higher than those obtained in horizontal orientation (Table 3).

TABLE 3. Specific extinction area for EPR cable burning with the assistance of the butane line burner.

Specific extinction area	Horizontal EPR cable	Vertical EPR cable
σ_{488} ($m^2.kg^{-1}$)	1128	1390
σ_{633} ($m^2.kg^{-1}$)	822	980
σ_{780} ($m^2.kg^{-1}$)	531	740

The experimental procedure used to burn fire-retarded electric cables brings in an external assistance, i.e. the flame of a burner. This flame gives rise to a dilution of the smoke plume, due to the resulting entrainment of air. It induces an upward flow which contributes mainly to the flow in the free stream section of the exhaust duct but does not participate to extinction (butane flame provides little if any smoke). Then, the influence of these joint (thermal and aerodynamic) effects on the conserved character of the specific extinction area must be considered. This has been done using a non fire-retarded material. Two sticks of PMMA were burnt, in horizontal orientation, with and without the assistance of the burner. The obtained values are reported in Table 4. It is seen that the discrepancy between the results with and without assistance is slight and within the experimental uncertainty. Thus, it can be concluded that the influence of the assisting flame on the specific extinction area is negligible.

TABLE 4. Influence of the assisting flame on the specific extinction area for a stick of PMMA.

Specific extinction area	PMMA sample alone	PMMA sample with flame burner assistance
σ_{488} ($m^2.kg^{-1}$)	270	255
σ_{633} ($m^2.kg^{-1}$)	189	186
σ_{780} ($m^2.kg^{-1}$)	156	140

From the results relative to EPR cable burning with assistance (Table 3) and to heating oil burning in the pan of 10 cm in diameter (Table 1 or 2), the relation (11) can be applied to calculate values for the extinction area relative to the smoke mixture. These values can be compared to the experimental ones. Such a comparison is made in Table 5 for the combustion of a vertical sample of EPR cable, and in Table 6 for the combustion of a horizontal sample of EPR cable.

TABLE 5. Summary of the results obtained with a pool of heating oil and a vertical sample of EPR cable burning in the flame of this pool.

Specific extinction area	Heating oil 10 cm pan	Vertical EPR cable with flame assistance	Mixture	Relation (11)
σ_{488} ($m^2.kg^{-1}$)	1464	1390	1340	1315
σ_{633} ($m^2.kg^{-1}$)	997	980	1065	1037
σ_{780} ($m^2.kg^{-1}$)	848	740	810	788

TABLE 6. Summary of the results obtained with a pool of heating oil and a horizontal sample of EPR cable burning in the flame of this pool.

Specific extinction area	Heating oil 10 cm pan	Horizontal EPR cable with flame assistance	Mixture	Relation (11)
σ_{488} ($m^2.kg^{-1}$)	1464	1128	1087	1192
σ_{633} ($m^2.kg^{-1}$)	997	822	855	938
σ_{780} ($m^2.kg^{-1}$)	848	531	680	731

It is seen that the calculated and experimental values are in agreement when the cable sample is oriented vertically. In contrast, when the sample is in horizontal position, it can be observed that the experimental values are noticeably lower than the calculated ones. It is assumed that, in this configuration, the sample constitutes, owing to its size, an obstacle to the upward flow and that the result is a significant blocking effect of the soot particles and a decrease of the mixture smoke concentration. In order to confirm this assumption, experiments were made introducing a sample of material generating little soot (stick of

PMMA) in place of the EPR sample, in the flame of the heating oil pool. In other tests of the same kind, the sample introduced in the flame was an inert material (steel cylinder). The smoke extinction coefficient measured during these tests revealed the same behaviour, as illustrated on the following Figure 5.

Figure 5 shows the evolution of the resulting extinction coefficient as a function of time, in the case of a PMMA sample. When the sample is introduced in the flame, the extinction coefficient decreases sharply. As soon as the sample is withdrawn, the value regains its initial level. The solid exhibiting a large surface comparatively to the pool fire, it effectively blocks a part of the soot generated by the flame.

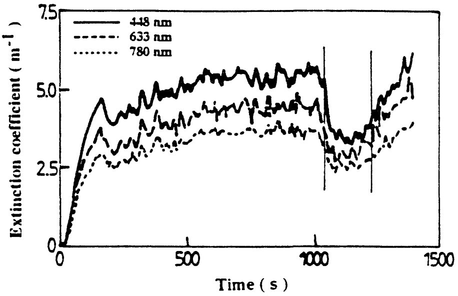


FIGURE 5. Evolution of the extinction coefficient as a function of time when a stick of PMMA is introduced in the flame of a heating oil pool and next withdrawn.

The above result shows that two materials burning, one engulfed in the flame of the second, may produce less smoke than if they burn separately. The consequence is a more or less marked diminution of the resulting amount of soot particles convected, depending greatly on the respective size and position of the two materials. To a lower extent, but noteworthy, is the possible additional effect of products as water vapour, carbon dioxide or monoxide, added to the stream by the material burning within the flame, which may be expected to influence the course of carbon production in this flame. A process of carbon particle oxidation with oxidants such those mentioned above, may compete with the two stages of soot formation processes: conversion of fuel to carbon nuclei and growth of carbon nuclei by a heterogeneous gas-solid reaction, and the propensity to liberate soot depends on the balance between these processes. Moreover, it is known that soot production is very sensitive to the conditions of dilution. The effect of dilution is further influenced by the dimension of the fire source. But, as regard this effect, there is still a need of specific studies.

CONCLUSION

In real fire situations, it often arises that several fire sources grow simultaneously due to spread to nearby objects by intense radiant heat transfer or direct flame impingement. Thus, to be in position to characterise the corresponding smoke mixtures produced is a problem of particular interest. In this prospect, studies have been conducted in laboratory-scale experiments. Although test of small size cannot model real situations, this facilitates the experiments, while it is anticipated that the results will be informative. Simple theoretical consideration has been used to establish the resulting specific extinction area for a mixture of smoke generated by various fire sources. This concept appears clearly as the more convenient quantity by which to attempt relevant correlation. Although the data scatter somewhat, the theoretical justification holds reasonably well. It has also been shown that the smoke generated by the burning of materials containing fire-retardants can be characterised in the same way, insofar as the assistance of a necessary heat supply (pilot flame or flame from another fire source) is provided. However, although the results of the present study provide valuable information, there is still a need to develop correlation able to predict full-scale performance.

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