

# HYDROCARBON AEROSOL EXPLOSION HAZARDS: A Review

P. J. BOWEN and L. R. J. CAMERON

*Cardiff School of Engineering, Division of Mechanical Engineering and Energy Studies, University of Wales, Cardiff, UK*

Potential explosion hazards arising from releases of natural gas are now reasonably well understood, but the hazards exhibited by two-phase liquid-vapour mixtures are often overlooked. A number of explosions believed to be two-phase in nature have occurred within the process industries, so emphasizing the need to investigate the underlying mechanisms which contribute to aerosol explosions. It has been predicted that an aerosol containing droplets within the so-called ‘transition’ range, typically between 7–15  $\mu\text{m}$ , may burn faster than an optimally mixed homogeneous vapour/air mixture. Other models predict that the burning velocity attained for a gaseous mixture is the maximum attainable. A few experimental publications discuss the generation and combustion of fuel aerosols, but have failed to examine the area of most interest. This paper reviews previous theoretical, experimental and computational approaches relating aerosol explosions, and presents a research strategy suitable for progression towards robust quantification of this category of explosion.

*Keywords: explosion; aerosol; hazard; review.*

## INTRODUCTION

This review paper collates the findings of previously unpublished work presented at United Kingdom Explosion Liaison Group (UKELG) meetings<sup>1,2</sup>, updated with recent experimental and theoretical research findings pertaining to the explosion hazard posed by hydrocarbon aerosols.

The potential explosion hazard posed by releases of flammable gases such as natural gas or propane are fully appreciated within the process industries. However, the hazards presented by releases of two-phase liquid-vapour mixtures (henceforth referred to as aerosol mixtures) are often underestimated. There are sufficient historical examples of incidents falling into this category of explosion<sup>3–6</sup> to warrant this position paper which outlines current understanding of mechanisms contributing to the generation of two-phase explosions, and presents a research strategy to allow flammable aerosols to be placed in their appropriate position in the hierarchy of explosive materials.

Explosion hazards from two-phase liquid fuel releases are likely to result from very dynamic events, such as the jet release of highly pressurized fuel or the release of superheated fuels, resulting in the formation of a hybrid, two-phase fuel-air mixture. Due to the highly transient nature of this type of fuel system prior to ignition, effects such as droplet evaporation and turbulence add considerable complexity to the analysis of the problem, but nevertheless need to be taken into account in any modelling approach or experimental validation programme.

The basic mechanisms contributing to overpressure generation in aerosol explosions are envisaged to be comparable to the corresponding vapour-cloud case, with turbulence-generated flame acceleration playing an influential role as outlined in the Schelkin mechanism<sup>7</sup>. A

mechanism based on that of Schelkin but revised to include the primary aerosol fuel effects is postulated in Figure 1. This helps focus on the various physical processes that need consideration when compiling an integrated modelling approach to quantifying this explosion hazard. The complexity in comparison with a single-phase explosion is now self-evident, primarily caused by the fact that the fuel characteristics ahead of the flamefront could be continually changing *during* the explosion. Moreover, the analogies with engine combustion are clear. Hence, not only is this of interest in explosion hazard analysis where conditions are uncontrolled, but the problem is also of relevance within relatively controlled combustion environments such as those in aeroengines or fuel-injected internal combustion engines, albeit under elevated temperatures and pressures.

As a starting point, if monodisperse aerosols are considered then one can simplify the problem if ‘worst-case’ conditions are adopted—a common approach in hazard analysis. Constituent droplet sizes should be between 5 and 20  $\mu\text{m}$ , as these are gravimetrically the most stable and likely to induce fastest burning—see later discussion. Clearly now stratification and ignition differences<sup>8</sup> are not relevant. Based upon Weber number criteria, droplet break-up would only become influential for fast explosions. Phase-change effects inducing variation in expansion ratio can be shown to be small, whilst radiative heating of droplets ahead of the flamefront and coalescence will move fuel characteristics away from ‘worst-case’ conditions, and so can also be eliminated. Variation of the external explosion characteristics will still be pertinent even in this simplified scenario, but this enigmatic phenomenon is a contemporary area of research even for single-phase explosions<sup>9</sup>. (The external explosion occurs outside a vent after the flame emerges from a semi-confined explosion, and tends to be a very fast

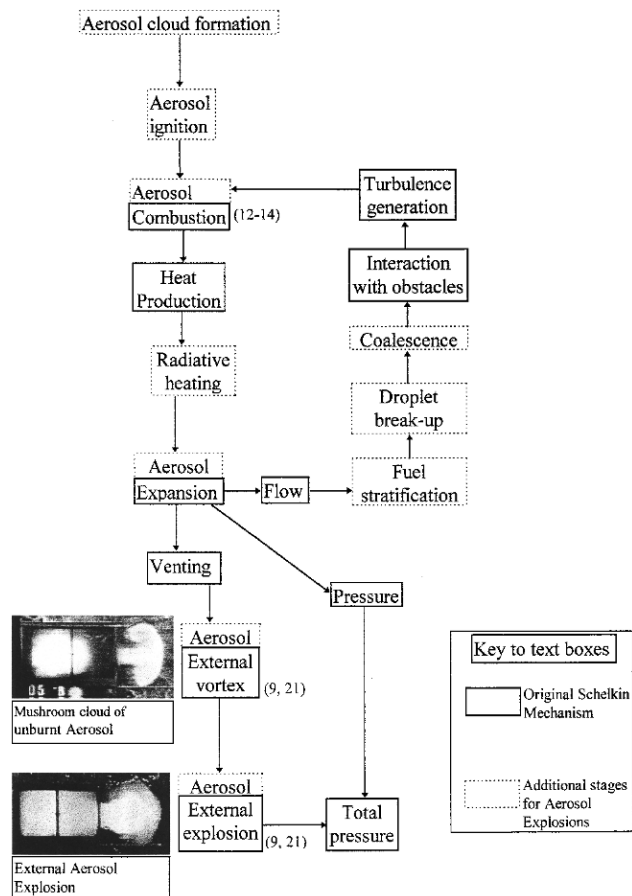


Figure 1. Mechanism for overpressure generation in fuel aerosol systems.

event due to the highly turbulent state of the fuel that has accumulated during the venting process.) Hence the primary difference between 'worst-case' scenarios for single- and two-phase vented explosions is the aerosol burning rate, which should be the basic building block underpinning future aerosol explosion models.

Explosion hazard models currently exist which allow predictions of overpressure and flame speed resulting from accidental gaseous explosions in typical process environments, where enclosure walls present restrictions to the expanding volume of burnt gas, and process equipment provides 'obstacles' which can induce flame acceleration. These models are invariably underpinned by databases of validation tests in scaled-down experimental facilities. Analogous models are not available for predicting the effects of two-phase explosions for two reasons: firstly, 'worst-case' aerosol fuel burning characteristics have yet to be established, and secondly there is no quality experimental data in scaled-down vented and/or 'congested' rigs against which codes can be validated.

There is speculation that two-phase hydrocarbon explosions may, under certain conditions, burn faster than homogeneous mixtures, and hence present a more severe hazard than that of an equivalent vapour cloud. Mainly theoretical studies have provoked this counter-intuitive hypothesis, although a couple of experimental studies have also tentatively suggested similar conclusions. These studies indicate that for monodisperse aerosol fuels within a so-called 'transition' range—typically somewhere

between 7 and 15  $\mu\text{m}$ —aerosol mixtures could have faster burning velocities than the fastest burning vapour cloud equivalent. Other researchers counter these predictions, arguing that the vapour limit provides the maximum burning rate attainable. Interestingly, these droplets are precisely the size that will remain suspended in the atmosphere after a large-scale aerosol release, their settling velocities being only of the order of  $10^{-2} \text{ m s}^{-1}$ . Larger particles will 'rain out' after only short periods of time, while most of those smaller will rapidly evaporate.

In light of previous experience trying to quantify the relative severity of aerosol explosions, experimental methods considered to be appropriate for future well-characterized experimental programmes are suggested, with a view to providing validation data suited to the various modelling approaches already proposed for quantifying aerosol burning rates and hence aerosol explosions.

## EXPERIMENTAL INVESTIGATIONS

The first significant advance concerning understanding of flame propagation through aerosol fuel mixtures was the classical treatise of Burgoyne and Cohen<sup>10</sup>. Monodisperse tetralin aerosols were produced in the particle size range 8–38  $\mu\text{m}$ , and subsequent flame propagation was characterized in a 'stationary-flame' experimental facility. It was noted that a transition in the character of the flame took place at about 10  $\mu\text{m}$ , below which the appearance of the flame was that of a pre-mixed gas-burner flame, where drops approaching the flamefront could be volatilized and mixed with air as quickly as the vapour formed could burn at the limit concentration. For larger drop sizes, the flame took the form of a brush-like spray of discrete burning drops, where drops burned diffusively in their own envelope of air and flame, one burning drop igniting adjacent ones, spreading combustion in a 'network' fashion. It was concluded that the burning velocity in an aerosol of 'large' drops is significantly higher than for a comparable aerosol of 'small' drops, although the method of aerosol generation precluded an independent study of fuel concentration and particle size effects. Assuming a temperature distribution across a tetralin flame and considering the dominant processes influencing evaporation, the rate of change of drop diameter was derived from which the critical drop diameter for homogeneous flame propagation was estimated to be about 14  $\mu\text{m}$ .

Several researchers including Mitzutani and Nakajima<sup>11</sup> investigated droplet burning rates in stationary flames during the early 1970s, but the most interesting developments concerned a series of novel experimental programmes considering flame propagation through fuel droplet, fuel vapour and air mixtures (Hayashi *et al.*<sup>12–14</sup>). Utilizing an aerosol generator similar in principle to Wilson's cloud chamber<sup>15</sup>, reasonably monosized ethanol and octane aerosol clouds were generated; the quantity of pre-vaporized fuel could be systematically varied. It was reported that the burning velocity of such heterogeneous mixtures could be greater than that of a homogeneous mixture of the same overall fuel-air ratio, provided that the droplet size was sufficiently 'large'. Combustion experiments were performed with droplet Sauter mean diameter (SMD) ranging between 4 and 30  $\mu\text{m}$ , and the flame structure produced was subsequently studied using

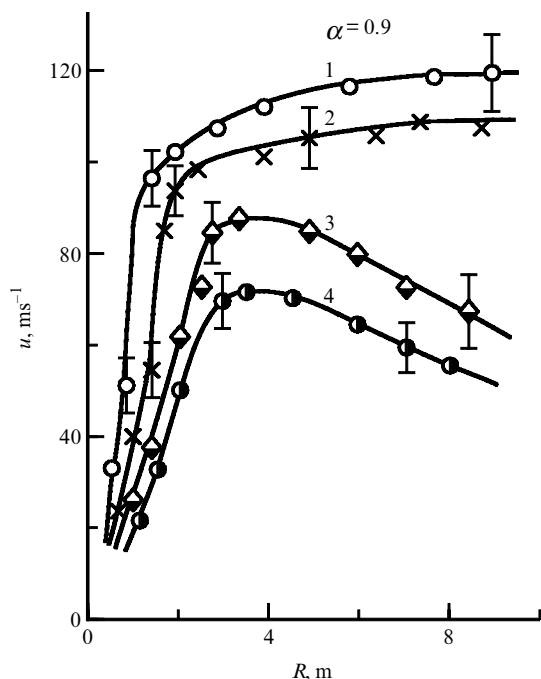


Figure 2. Apparent velocity of flame front in fuel aerosols ( $\alpha$  denotes the statistical reliability): 1—fuel oil, 2—diesel oil, 3—kerosene, 4—gasoline. Reproduced from Reference 20. Reprinted by permission of Plenum Publishing Corporation, 1989.

Schlieren and high-speed photography. (Sauter mean diameter is the diameter of a droplet whose surface-to-volume ratio is equal to that of the entire spray.) Flame structures observed during propagation through heterogeneous mixtures containing droplets between 4 and 7  $\mu\text{m}$  were found to be smooth in appearance, as exhibited in homogeneous combustion. It was also noted that the flame structure became increasingly 'wrinkled' as the droplet size was increased from this lower limit.

The authors concluded that the burning velocity in each instance was governed by the fuel-air ratio in the gas phase ahead of the flame front. This ratio consists of contributions from two different sources: vaporisation of droplets being heated just ahead of the flamefront, and the fuel vapour in the two-phase mixture pre-ignition. The contribution from the former source is obviously dependent upon the mean droplet diameter. Chan and Jou<sup>16</sup> and Chan and Wu<sup>17</sup> reported similar findings, but were able to explain their observations in terms of a relative shift in the 'effective' equivalence ratio corresponding to fastest burning in the mixture.

Ballal and Lefebvre<sup>18</sup> subsequently investigated aerosol flame propagation using Fuller's method<sup>19</sup> of utilizing acoustic devices to create flat flames in tubes. Aerosol fuel mixtures of iso-octane, diesel oil and heavy fuel oil were prepared for various equivalence ratios and several mean drop diameters (100, 60 and 30  $\mu\text{m}$ ). A vaporizer and atomizer were used to prepare the required droplet/vapour mixture. Their results showed that the burning velocity of mists was appreciably enhanced by a reduction in drop size to 30  $\mu\text{m}$ , and that the presence of fuel vapour in a multi-droplet environment was beneficial to flame propagation. As the smallest particles considered were only 30  $\mu\text{m}$ , effects within the 'transition' particle size range were not investigated and, as expected, the fastest flames recorded were less than the burning velocity of the vapour.

The first investigation to attempt aerosol deflagration experiments at large scale was conducted by a Ukrainian group (Kopyt *et al.*<sup>20</sup>). 600  $\text{m}^3$  aerosol fuel clouds dispersed in the open atmosphere were ignited centrally. Deflagrations through gasoline, kerosene, diesel oil and fuel oil clouds were initiated in order to determine the effect of fuel type upon flame speed. Droplet size distributions created by the super-heated fuel atomizer were quoted to be within the range 18–28  $\mu\text{m}$ , depending on the particular fuel being tested. High speed photography was used to visualize the aerosol formation, cloud uniformity and measure flame speed. The large clouds of aerosol fuel supported flame-speeds of up to 100  $\text{m s}^{-1}$ , the faster flame speeds occurring for the 'heavier' fuels (Figure 2). However, the method of aerosol generation would have undoubtedly created significant pre-ignition turbulence, a high-energy ignition source was used, and there is some doubt with regard to the authors' definition of 'flame speed'. These factors cast uncertainty on the validity of the results of this programme. The authors claim that heat transfer via radiation played a significant role in the combustion of the large aerosol fuel clouds, preheating the aerosol ahead of the cloud. Radiative heating is usually ignored in modelling studies of aerosol flame propagation, but accidentally-released industrial fuel aerosol clouds would be of this scale or larger, so radiative heat transfer would be more relevant in a real accident.

Medium-scale aerosol tests have been recently reported (Bowen *et al.*<sup>21</sup>) where the aim was to examine explosion hazards posed by aerosols within the 'transition' particle size range. Aerosols were generated using 'fine' spray atomizers in an uncongested rectangular box approximately 2.5  $\text{m}^3$  in volume, containing a single vent in one of the end faces. A low-energy ignition source was devised to initiate reaction, and pre-ignition turbulence was accounted for within the experimental methodology adopted to facilitate a comparative study of fuel mixtures.

Hydrophones positioned both inside and outside the chamber measured explosion overpressure, and flame position was recorded using high-speed photography. Initial tests of propane/air mixtures were conducted to provide a benchmark for comparison with aerosol tests, which comprised kerosene, decane, heptane, propane/heptane and propane/kerosene mixtures. For the purposes of explosion hazard assessment, it was concluded that low volatility fuel aerosol explosions (decane and kerosene) were comparable to very lean methane explosions at this scale (Figure 3). Note this conclusion is in direct conflict with the larger scale study of Kopyt *et al.*<sup>20</sup>. By contrast, volatile fuel aerosols, such as those containing heptane, tended to induce faster flame acceleration than propane/air aerosol explosions (Figure 3). The general conclusions of this research are consistent with an earlier unpublished multi-industry research programme conducted by TNO (The Netherlands) and CMR (Norway) (MEXOS, 1992).

## THEORETICAL MODELS

Motivated by the intriguing experiments of Burgoyne and Cohen<sup>10</sup>, Williams<sup>22</sup> presented the first theoretical model of flame propagation through heterogeneous mixtures. Considering one-step kinetics, and dividing the process into 'vaporization' and 'reaction' zones, he derived a critical rate ratio which determines whether homogeneous or

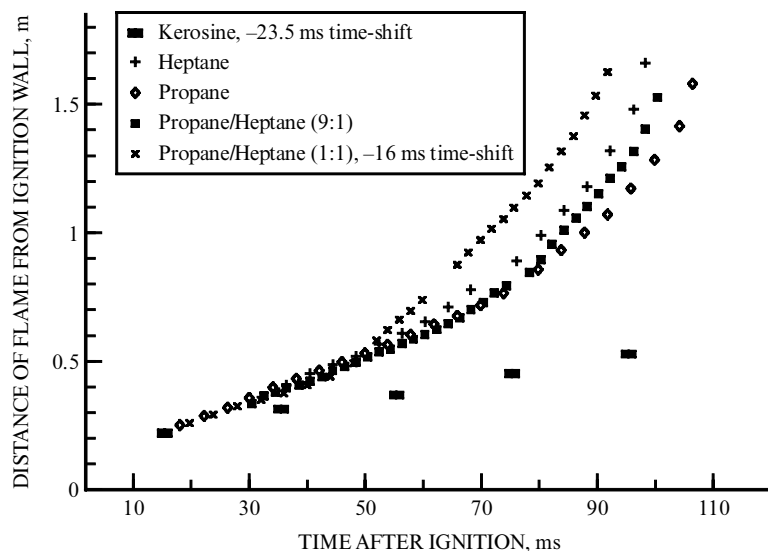


Figure 3. Distance/time-of-arrival data for optimized propane and various aerosol-hybrid deflagrations. Reproduced from Reference 8. Reprinted by permission of OPA (Overseas Publishing Association) Amsterdam B.V. Published under licence under the Gordon and Breach Science Publishers imprint.

heterogeneous combustion dominates flame propagation. From this condition he was able to estimate the lower critical particle size below which homogeneous combustion prevails. His estimated value of  $9\ \mu\text{m}$  was almost identical to that measured by Burgoyne and Cohen<sup>10</sup>. He also derived an analytical expression for the heterogeneous burning velocity for aerosols containing droplets larger than those in the transition range. However, comparing the predicted results with experiment showed about a 30% underprediction. He provided analysis of the equations governing homogeneous flame propagation under various limiting conditions, but concluded, 'The difficult problem of predicting the burning velocity in the transition region, where both homogeneous and heterogeneous processes occur simultaneously, remains unresolved'.

Ballal and Lefebvre<sup>18</sup> provided a theoretical interpretation of aerosol flame propagation by considering the quench time of the reaction zone to be the sum of the evaporation and chemical reaction times. Deriving expressions for each of these characteristic times allows an expression for the flame thickness to be determined, from which the following expression for the aerosol burning velocity results:

$$S = \alpha_g \left[ \frac{C_3^3(1-\Omega)\rho_f D_{32}^2}{8C_1\rho_g \log(1+B)} + \frac{\alpha_g^2}{S_L^2} \right]^{-0.5} \quad (1)$$

Clearly this predicts a monotonic decrease in aerosol burning velocity from the vapour limit ( $S_L$ ), i.e. the homogeneous combustion limit (Figure 4). The model is validated by the data described in the experimental section, but only for aerosols with particle size greater than  $30\ \mu\text{m}$ .

A variation of this model was provided a few years later by Polymeropoulos<sup>23</sup>. Providing an energy balance for the reaction zone and considering droplet vaporization up until time of droplet ignition results in the following expression for aerosol burning velocity in terms of the vapour equivalence ratio, droplet size at ignition, thermal diffusivity, densities of vapour and liquid, droplet size in the unburned mixture and the mass transfer constant for droplet

combustion:

$$S^2 = \left\{ \frac{D_i^5(1-\Omega)}{12\alpha^2 D_u^3 \left(\frac{\rho}{\rho_f}\right) \ln(1+B)} + \frac{1}{S_L^2} \left[ 1 - \frac{D_i^3}{D_u^3} (1-\Omega) \right] \right\}^{-1} \quad (2)$$

Knowledge of the droplet size at ignition is required to allow a solution to this equation, and this is provided using the theory of Law and Chung<sup>24</sup>. It is noted that for the homogeneous combustion regime ( $D_u = D_i$ ) equation (2) is comparable to equation (1) if the heat release is dominated by droplet vaporization (when  $\Omega$  is sufficiently small and for sufficiently large droplets). When  $D_u < 30\ \mu\text{m}$ , whereas equation (1) predicts the maximum velocity at the limiting vapour state, equation (2) predicts peaks of burning velocity before decreasing to the  $S_L$  limit (Figure 5). Largest burning velocities were predicted for the mixture with smallest upstream vapour fraction.

Burning velocity versus droplet diameter relationships for both iso-octane and medium diesel-oil at various fuel fractions are presented in Figures 4 and 5. Comparing the predictions of the proposed model against the results obtained by Ballal and Lefebvre is interesting, emphasizing the burning velocity promotion due to the presence of aerosol droplets.

One-dimensional unsteady two-phase computational fluid dynamic (CFD) codes for studying flame propagation within closed vessels were developed during the 1980s by Sirignano, Aggarwal and co-workers. Early derivations were based on Eulerian-Eulerian treatment of the two systems of equations representing each phase, but whilst the continuum approach is appropriate for the gaseous phase, it was difficult to justify this for the discrete droplet phase. Later codes<sup>25</sup> developed a hybrid Eulerian-Lagrangian approach primarily to avoid problems of numerical diffusion. Semi-empirical correlations were used for some of the droplet effects such as drag and vaporization. Interaction between phases was treated by using appropriate interpolation and source distribution terms, and finite difference

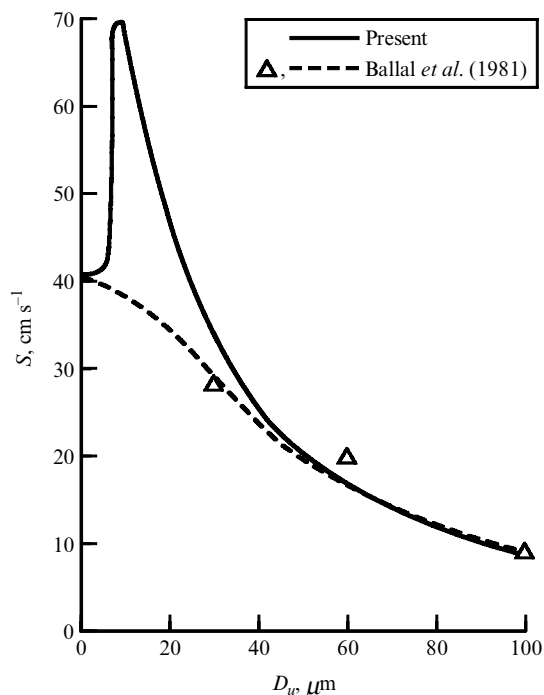


Figure 4. Previous predictions of variation in laminar burning velocity with droplet diameter. Reproduced from Reference 23. Reprinted by permission of Gordon and Breach Science Publishers Inc, 1984.

techniques were employed as the numerical solver for the resultant system of equations. Grid sizes used were small compared to the distance between droplets, but coarse compared with droplet diameter. A one-step chemical kinetics model was employed. Earlier versions of the codes<sup>26,27</sup> were unable to model flame propagation to the end of the combustor due to the computational expense of finer grid sizes in the regions of high gradients, whereas later versions were capable of modelling the flame propagation to the end of the combustor using non-uniform grid-spacing and adaptive time step. The code was also extended to include multi-component fuel droplets. Radiative effects and droplet-droplet interactions were neglected.

Fuels relevant to the automotive industry, such as octane and decane, were considered. Parametric studies undertaken included the effects of overall equivalence ratio, initial particle size and fuel volatility. These studies showed that flame propagation seemed to increase with the volatility of the fuel. For aerosols with droplet size within the transition range, increases in flame propagation rate above the premixed limit for a particular fuel were noted, but only for *off*-stoichiometric mixtures. It does not appear that a systematic study of varying overall equivalence ratio for droplet size fixed within the transition range has been undertaken. The difference in motion between the droplets and vapour induces stratification in the mixture, which results in the flame exhibiting simultaneously both diffusion and premixed-like character. The premixed character becomes more important as fuel volatility increases or initial droplet size decreases. No indication of individual droplet burning was noted, with nearly all the vaporization occurring either behind or ahead of the propagating flame.

Lin and Sheu<sup>28</sup> presented a theory of laminar flame propagation using matched asymptotics initially for *off*-stoichiometric dilute sprays, and then for near-stoichio-

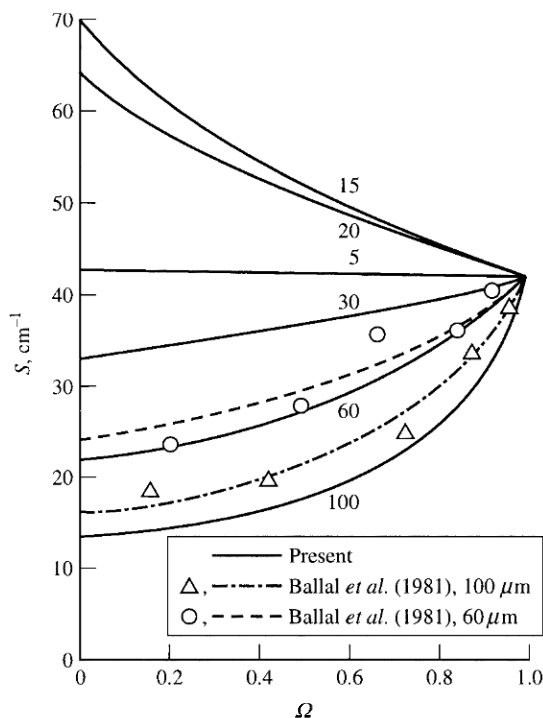


Figure 5. Previous model showing effect of  $\Omega$  on flame speed in iso-octane fuel sprays. Numbers denote droplet Sauter mean diameters. Reproduced from Reference 23. Reprinted by permission of Gordon and Breach Science Publishers Inc, 1984.

metric sprays in the limit of large activation energy. For sample calculations, octane was chosen as the fuel under initial conditions of atmospheric pressure and 75°C. The most interesting aspects of the predictions in the context of this paper are in the cases of fuel-rich sprays. The peak burning velocity for the fuel vapour at this temperature is about 73 cm s<sup>-1</sup> at a total equivalence ratio of 1.15. For constituent droplets in the sub-20 micron range, and for a liquid fuel mass fraction of 0.13, burning velocities of 78 cm s<sup>-1</sup> are predicted. For greater or lesser liquid fuel mass fractions the peak burning velocity decreases. This burning velocity enhancement is further accentuated for a total equivalence ratio of 1.35. Here peak velocities of 86 cm s<sup>-1</sup> are predicted—some 20% burning velocity enhancement—when the liquid fuel mass fraction is 0.24. Again for values of liquid loading away from this optimum the burning velocity prediction decreases, but remains in excess of the maximum vapour flame speed of 73 cm s<sup>-1</sup> when the liquid mass fraction is less than 0.24. No experimental data is available to appraise these predictions, but the authors state that the results are in agreement with the flame propagation mechanisms proposed by previous researchers (discussed earlier in this paper) and in qualitative agreement with the observations of Hayashi *et al.*<sup>12-14</sup>.

Greenberg *et al.*<sup>29-32</sup> have recently analysed and predicted burning velocities for spray flames. However, their results had always predicted velocities less than that of an equivalent single-phase gaseous premixed flame. In a recent note<sup>33</sup>, the authors model the mechanism of flame wrinkling first alluded to by Hayashi *et al.*<sup>13</sup> to induce relatively small burning velocity enhancement, which would be consistent with the limited experimental evidence to date. The simple model introduces a methodology

to account for the 'ruggedness' induced within the flame front due to the small-scale mixture heterogeneity. Droplets of size  $D$  are presumed to deform the flame surface into hemispheres of radius  $b(D)D$ . The increase in surface area of the flame front due to distortion-causing droplets of diameter  $D$  is then:

$$\Delta s = N\pi b^2 D^2/4 \quad (3)$$

where  $N$  is the number of droplets of diameter  $D$  per unit area responsible for flame-front wrinkling:

$$N = a(D)m\rho/(\pi\rho_l D^3/6) \quad (4)$$

Simplifying the problem by taking  $a(D)$  and  $b(D)$  as constants, and integrating with respect to  $D$  over all the diameters of droplets in the spray, the following equation for burning velocity of the 'rugged' aerosol flame can be obtained. It is presented in a form which suggests an analogy with turbulent burning:

$$S_{rugged} = S_L(1 + \Lambda G_{df}) \quad (5)$$

If validated, this modification would obviously be straightforward to implement in existing explosion models.  $S_L$  is the burning velocity of the planar spray flame,  $G_{df}$  is the (non-dimensional) mass fractional flux of droplets at the undisturbed flamefront, and  $\Lambda$  for a particular fuel is a constant. The implication of this equation is that as the initial quantity of droplets supplied to the premixture and/or their diameters increase, the quantity of droplets that will not vaporize before reaching the flame front may increase, hence increasing the flame-front distortion and the burning velocity.

Length scales associated with flame wrinkling due to spatial distribution of droplets within the transition range at stoichiometric concentration are  $O(10^{-4})$ m. In a pseudo-laminar droplet flame as in the experiments of Hayashi *et al.*<sup>12-14</sup>, this will be the only 'flame wrinkling' length scale present, and the results of Greenberg *et al.*<sup>30-33</sup> show using a simple geometrical analysis that its influence on burning rate is only small. In the spirit of Greenberg *et al.*<sup>30-33</sup>—that is, continuing the analogy of flame wrinkling due to aerosol heterogeneity and turbulence-induced wrinkling—in a fully turbulent environment as encountered in most practical applications, this scale needs to be taken into consideration with respect to the inner cut-off limit of scales that influence flame wrinkling. However, in a fully turbulent environment the mechanism of flame wrinkling due to aerosol heterogeneity is likely to provide negligible contribution to overall flame acceleration, and if this is the case, flame acceleration will be dominated by the interaction between turbulence and fuel volatility in aerosol explosions. Certainly this would be consistent with existing medium-scale experimental evidence<sup>8,21</sup>, but not with the large-scale experiments of Kopyt *et al.*<sup>20</sup>.

If this interpretation is correct, and existing experimental data valid, then it is apparent that aerosol explosions scale in a different manner to single-phase explosions, perhaps due to the varying influence of different processes in the proposed 'aerosol' version of the Schelkin mechanism in Figure 1 as scale increases.

Gauthier and Bardon<sup>34</sup> considered a general phenomenological flame propagation model for fuel droplet, particle and vapour mixtures in air. The model hypothesis is that the heterogeneous burning velocity is equal to that of a

homogeneous mixture of 'effective' equivalence ratio derived as the sum of the original fuel vapour present and that produced by evaporation as droplets pass through the flame front. The 'effective' equivalence ratio is also dependent upon the burning rate, and hence an iterative procedure is required to provide a solution for the aerosol burning velocity. However, from a 'hazard' perspective, the model can not predict a burning velocity in excess of the vapour limit. The authors continue to introduce effects of multi-component fuels and turbulence enhancement of burning rate.

## FUTURE AEROSOL EXPLOSION VALIDATION STUDIES

As seen from previous sections, several modelling approaches have been proposed for predicting flame propagation through aerosol mixtures. It is essential now that benchmark experimental data are provided so that these models can be systematically appraised. The difficulty in providing validation data for explosions in aerosol fuel systems is that several fuel-related characteristics need to be quantified and controlled before ignition due to the two-phase nature of the system. By contrast, studies of flame propagation in vapour clouds only require homogeneity of the mixture and quantification of the fuel vapour concentration. Hence, an adequate methodology for fuel-air mixing and on-line sampling of the mixture passing through a suitable gas-analysis sensor (e.g., infrared gas analyser for methane/air) is sufficient to characterize the vapour fuel system prior to ignition. For aerosol fuel clouds, again homogeneity needs to be established, but in addition (i) particle size, (ii) total equivalence ratio (including fuel vapour and fuel contained in droplets), (iii) droplet equivalence ratio and (iv) pre-ignition turbulence ideally need to be quantified. As aerosol systems are transient by nature—evaporation, settling and interaction with boundaries ensure that the fuel characteristics are continually varying—it is important that any characterization technique utilised is capable of temporal resolution. Finally, the ability to vary these four primary fuel parameters is required for a complete study, i.e. a robust, repeatable method of atomization and fuel mixing is required.

Given these very stringent criteria, it is not surprising that in the authors' opinion no satisfactory experimental study at any scale has been conducted to date. However, the advances of temporally resolved radiation-based techniques means that a systematic experimental study should presently be possible at least at laboratory scale. The following sections summarize the experimental techniques currently available or under development which seem suited to addressing the four primary characterization requirements stated above. First, methodologies appropriate for aerosol generation are summarized.

## METHODS SUITABLE FOR AEROSOL GENERATION

In view of simplifying the analysis of aerosol combustion, the distribution of droplet sizes present in the aerosol system should be as narrow as possible, i.e. having a high degree of monodispersity. A variety of methods have been used to

generate aerosols<sup>12,13,35</sup> for aerosol flame propagation studies. Nozzles, such as those employed in gasoline and diesel internal combustion engines, are capable of creating a fine, turbulent mist with droplet sizes ranging typically from 5–40  $\mu\text{m}$ . Nozzles, therefore, are not ideally suited to aerosol burning velocity studies due to the polydisperse nature and turbulent conditions of the spray produced.

For larger scale studies, the rapid release of superheated fuels can also lead to the formation of a very large aerosol cloud of fine droplets<sup>20</sup>, but droplet size, cloud size and turbulence are difficult to control.

The apparatus used by Hayashi *et al.*<sup>12,13</sup> is based upon the principle of operation of the Wilson cloud chamber<sup>15</sup>. A near monodisperse aerosol is formed when a saturated fuel vapour/air mixture is expanded. Aerosols generated in this way can be used for flame propagation rate investigations in the transition range, as the droplet size present in the aerosol cloud can be controlled with a high degree of accuracy.

### PARTICLE SIZE

One of the main concerns regarding hazards associated with aerosol fuel releases is how the flame propagates when constituent particles are in the 'transition' range<sup>10,12–14,28</sup>. Hence, the capability to measure droplet sizes across the transition range is imperative in quantitative experimental studies.

One of the more popular commercially available systems for particle sizing is the Malvern Particle Sizer. This uses a low-powered collimated laser beam and utilizes the diffraction properties of light passing through media of different refractive indices. Temporal resolution is afforded in the more recent Malvern MasterSizer X model, where the number of laser 'sweeps' can be controlled by the user. Droplets diffract the passing laser radiation, the angle of diffraction being related to the size of droplet. In principle, droplets in the range 0.1–2000  $\mu\text{m}$  can be measured, but for a particular experiment, considered choice of the range lens employed is required, as a dynamic range of only about 200 is available. Furthermore, care is required in the experimental design to avoid the so-called 'vignetting' effect. Vignetting occurs when light scattered from particles is not correctly resolved by the collection optics. It can be avoided by ensuring that the spray is within the appropriate working distance of the collection lens. Vignetting affects sizing measurements by biasing the size distribution towards larger mean droplet sizes. Recent calibration studies by the authors have indicated that problems of vignetting can be avoided for particles around the transition range for small-scale combustors using standard optics. For studies at larger scale, large-scale non-standard 'Fourier-Transform' collector lenses and telescopic beam projectors are required to avoid vignetting<sup>8</sup>.

Phase Doppler Anemometry (PDA) is another popular non-intrusive technique that can be employed to measure droplet diameters as well as three components of droplet velocity simultaneously. To obtain a characteristic size of a droplet population, thousands of particles need to be measured over a finite time period, the duration of which is dependent upon the data collection rate, which in turn depends upon the aerosol cloud characteristics. Hence, PDA is not considered an ideal technique for this application, due to the requirement of temporal resolution. Details of the principle

operation may be found elsewhere. Instantaneous laser-sheet particle sizing is also under development, and would seem suitable for this application after further refinement and calibration.

Shell Research has recently developed an imaging technique ILIDS (Interferometric Laser Imaging for Droplet Sizing) which allows temporal quantification of droplet fields in SI engines. Glover and Bowen<sup>36</sup> identified ILIDS as a possible droplet sizing technique for sizing controlled explosion assessment. When out-of-focus images of scattered light from droplets are recorded on film, interference occurs between reflected and first order internally refracted rays. It can be shown that for specific receiving angles, the relationship between particle diameter and number of fringes is linear. Hence, by using an automated image analysis system to identify each droplet interferogram and counting the fringes within, instantaneous droplet size distributions can be generated. This technique may well be suited to aerosol flame propagation studies, depending on the spray density limitations of the technique which have yet to be established.

### PRE-IGNITION TURBULENCE

Pre-ignition turbulence is likely to occur in most experimental studies of aerosol flame propagation studies to a greater or lesser degree depending upon the atomization technique employed.

Particle Image Velocimetry (PIV) is capable of allowing almost instantaneous visualization of any turbulent eddies present. A double pulse of an Nd:YAG laser sheet with suitable time separation between pulses can provide images of particle seeding or droplets within the flowfield. Using auto- or cross-correlation techniques, so long as the particles remain within the sheet, droplet velocity vectors can be derived knowing the time separation between pulses.

In this application PIV would provide quasi-instantaneous two-dimensional velocity flowfield and a measure of how quiescent the mixture is pre-ignition. In order to rigorously quantify  $u'$  throughout the explosion chamber many thousands of images would have to be taken in order to observe velocity fluctuations with time at discrete points.

As for the PDA, Laser Doppler Anemometry (LDA) relies on characterization of Doppler bursts as individual droplets pass through an optical control volume. LDA could be used as a measure of the transient decay of pre-ignition turbulence, but again as a single particle counter it also has temporal limitations.

### FUEL/AIR CHARACTERIZATION

Measurements of liquid, vapour and overall fuel:air ratios for aerosol mixtures are other important parameters for model validation purposes. A method developed by Shell Research Ltd. based on the absorption of infrared radiation has been shown capable of providing temporally-resolved fuel vapour concentration measurements in a heterogeneous fuel mixture<sup>37</sup>. As with the diffraction-based particle-sizing technique, the methodology uses a line-of-sight approach and relies upon the fact that carbon-hydrogen bonds absorb near infrared radiation (NIR) at certain wavelengths. A collimated infrared beam passing through

a two-phase hybrid fuel mixture is attenuated by (i) extinction due to liquid phase droplets and (ii) losses at the windows, which are usually sapphire or quartz to minimize beam attenuation. The vapour phase concentration is determined by making a simultaneous measurement at a second wavelength where there is no vapour phase absorption, and hence readings at this wavelength are solely due to attenuation.

Another technique continually being developed for fuel concentration measurements in combustion applications is Laser-induced Fluorescence (LIF). Three strategies are available for the fluorescent imaging of sprays: natural fluorescence from the fuel, fluorescence from a dopant molecule and from exciplex-based fluorescence. Both dopant and exciplex (*excited complex*) molecules are carefully tailored chemical additives that fluoresce at a known frequency. The added advantage of using exciplex-based fluorescence is that the fuel vapour emits a substantially different wavelength from that of the liquid fuel. The exciplex methodology would appear to be the most appropriate for this application as it discriminates between liquid and vapour fuel concentrations.

Two dopants need to be carefully selected and matched to evaporation characteristics of the particular fuel being considered. Dopant concentrations are selected such that two 'excited' molecules emit radiation at different wavelengths, one corresponding to the vapour and the other corresponding to the liquid fuel phase. Imaging through appropriate filters allows planar quantification of fuel in each phase. Fluorescent intensity is related to the fuel concentration, so that with appropriate calibration, quantitative results may be obtained. The technique is still under development for engine applications, contemporary problems including the quenching of vapour phase fluorescence in oxygen environments, and complications arising under conditions of elevated temperature and pressure. Only the former limitation would need consideration for this application.

## CONCLUSIONS

The most hazardous aerosol fuel clouds in the context of explosion assessment happen to contain droplet sizes which persist longest in large-scale releases, and hence are the clouds most likely to be encountered in industrial accidents. Burning velocity is the variable that will have most influence in determining the severity of an accidental aerosol explosion. Several models exist for predicting laminar burning rates through aerosol fuel clouds. They all agree that for particle sizes greater than 30  $\mu\text{m}$  the aerosol burning velocity cannot exceed the equivalent vapour burning velocity, as the flame propagates in the so-called 'heterogeneous' mode. This prediction is validated by several sets of experimental data at laboratory scale.

For aerosol fuel systems where the constituent particle size is between 7 and 15  $\mu\text{m}$ , (the so-called droplet transition range), there are inconsistencies in the various model predictions. Some models predict *significant* enhancement in laminar burning rate for particle sizes between the heterogeneous and homogeneous limit—where droplets completely evaporate in the preheat zone. However, no reliable data exists against which such predictions can be appraised.

Various laser diagnostic techniques have matured enough now to allow quantification of the influential fuel parameters in laboratory-scale investigations. There is a need to provide reliable benchmark data in small, well-controlled facilities before the complexities of two-phase explosions in industrial environments can be considered, or explosion models enhanced to include two-phase effects. This benchmark data should be appraised against detailed numerical simulations; such CFD models have already been developed.

Complications arising in real incidents include the effects of turbulence on aerosol flame propagation, droplet breakup and/or coalescence due to the explosion-induced flow field, polydispersity of the droplet cloud and effects of radiative heating of particles ahead of the flame front. Probably as a result of these and other secondary mechanisms, there are further inconsistencies between the conclusions of the few larger-scale two-phase explosion studies to date, but attempts to interpret the discrepancies are exacerbated by lack of experimental characterization detail and knowledge of the fundamental governing processes. Very high flame speeds in large-scale, highly turbulent releases of low-volatility aerosol fuel have been reported which would be consistent with enhanced burning mechanisms prevailing. However, recent medium-scale studies in a vented enclosure do not show evidence of significant increases in burning rate over that of the equivalent vapour mixture. Symbiotic studies between the combustion 'hazard' and 'engine' fraternities will aid understanding as the underlying problems are common.

It is recommended that in explosion hazard assessment, the simplistic current practice of modelling condensate fuels as propane vapour be continued until further research provides us with the information required to model aerosol effects within hydrocarbon explosions more rigorously.

## NOMENCLATURE

$a$	function used by Greenberg to measure the range in which droplets affect the flame front
$b$	function used by Greenberg to determine distortion of flame front due to droplets
$B$	mass transfer number
$C_p$	specific heat at constant pressure
$C_1$	$D_{20}/D_{32}$
$C_3$	$D_{30}/D_{32}$
$D$	droplet diameter
$D_{20}$	mean surface area diameter
$D_{30}$	mean volume diameter
$D_{32}$	Sauter mean diameter
$G_{df}$	nondimensional mass fraction
$m$	distribution function
$N$	total droplet number
$\Delta S$	increase in flame surface area
$S$	burning velocity of the two-phase mixture
$S_L$	burning velocity of the homogeneous mixture
$u_{rugged}$	burning velocity of 'rugged' flame
$u_{flat}$	burning velocity of planar premixed spray flame
$\alpha$	thermal diffusivity
$k$	thermal conductivity
$\Lambda$	equal to $(3/2)ab^2(\rho/\rho_l)$
$\Omega$	upstream fraction of total fuel in vapour form
$\rho$	mixture density
$\rho_l$	liquid fuel density

### Subscripts

$f$  fuel



g gaseous, or liquid fuel/vapour/air mixtures.  
 i ignition condition  
 u upstream condition

## REFERENCES

1. Bull, D.C. and Bowen, P.J., 1991, Explosions in two-phase mixtures, keynote address, *United Kingdom Explosion Liaison Group, Imperial College*.
2. Bowen, P.J., 1996, Trends in explosion research: Mist explosions, *United Kingdom Explosion Liaison Group, Aberystwyth*.
3. *The Flixborough Disaster, Report of the Court of Inquiry*, 1975 (HMSO, London).
4. The Hon. Lord Cullen, November 1990, *The Public Enquiry into the Piper Alpha Disaster (The Cullen Report)*, 2 volumes (HMSO, London).
5. Slater, D.H., 6 May 1978, *Chemistry and Industry*, 296–302.
6. Guban, K., 1979, *Unconfined Vapour Cloud Explosions* (Institution of Chemical Engineers, Rugby/Godwin, London).
7. Bimson, S.J., Bull, D.C., Cresswell, T.M., Marks, P.R., Masters, A.P., Prothero, A., Puttock, J.S., Rowson, J.J. and Samuels, B., 1993, *14<sup>th</sup> International Colloquium on the Dynamics of Explosions and Reactive Systems, 1–6 August, Coimbra, Portugal*.
8. Bowen, P.J. and Shirvill, L.C., 1994, *J Loss Prev Proc Ind*, 7 (3): 233–241.
9. Marsano, S., Bowen, P.J. and O'Doherty, T., 1998, *Proceedings of 13<sup>th</sup> Australasian Fluid Mechanics Conference, 13–18 December, Monash University, Melbourne, Australia*.
10. Burgoyne, J.H. and Cohen, L., 1954, *Proc of the Royal Society of London Academy*, No. 225, 375–392.
11. Mizutani, Y. and Nakajima, A., 1973, *Combust Flame*, 20 (3): 343–350.
12. Hayashi, S. and Kumagai, S., 1975, *15<sup>th</sup> Symposium (Int) on Combustion*, No. 6, 479–493.
13. Hayashi, S., Kumagai, S. and Sakai, T., 1976, *Combust Sci Technol*, 15: 169–177.
14. Hayashi, S. and Kumagai, S., 1980, *18<sup>th</sup> Symp (Int) on Combustion*, No. 19, 361–367.
15. Wilson, C.T.R., 1897, Condensation of water vapour in the presence of dust-free air and other gases, *Proc of the Royal Society of London*.
16. Chan, K. and Jou, C., 1988, *Fuel*, 67: 1223–1227.
17. Chan, K. and Wu, S., 1989, *Fuel*, 68: 139–144.
18. Ballal, D.R. and Lefebvre, A.H., 1981, *18<sup>th</sup> Symp on Combustion*, No. 19, 231–328.
19. Fuller, L.E., Parks, D.J. and Fletcher, E.A., 1969, *Combust Flame*, 13 (1): 455–460.
20. Kopyt, N.K., Struchaev, A.I., Krasnoshchikov, Y.I., Rogov, N.K. and Shamshev, K.N., 1989, *Combustion, Explosion and Shock Waves*, 25 (3): 279–285.
21. Bowen, P.J., Bull, D.C., Prothero, A. and Rowson, J.J., 1997, *Combust Sci Technol*, 130: 25–47.
22. Williams, F.A., 1960, *Progress in Astronautics and Rocketry*, No. 2, 229–264.
23. Polymeropoulos, C.E., 1984, *Combust Sci Technol*, 40: 217–232.
24. Law, C.K. and Chung, S.H., 1980, *Combust Sci Technol*, 22: 17–26.
25. Continillo, G. and Sirignano, W.A., 1988, *22<sup>nd</sup> Symp (Int) on Combustion, 1941–1949* (The Combustion Institute).
26. Aggarwal, S.K. and Sirignano, W.A., 1985, *Combust Flame*, 62 (1): 69–84.
27. Seth, B., Aggarwal, S.K. and Sirignano, W.A., 1980, *Combust Flame*, 39 (1): 149–168.
28. Lin, T.H. and Sheu, Y.Y., 1991, *Combust Flame*, 84 (3–4): 333–342.
29. Silverman, I., Greenberg, J.B. and Tambour, Y., 1991, *SIAM J Appl Math*, No. 51, 1284–1303.
30. Silverman, I., Greenberg, J.B. and Tambour, Y., 1992, *Atomisation and Sprays*, 2: 193–224.
31. Silverman, I., Greenberg, J.B. and Tambour, Y., 1993, *Combust Flame*, 93 (1): 97–118.
32. Greenberg, J.B., Silverman, I. and Tambour, Y., 1996, *Combust Flame*, 104 (3): 358–368.
33. Greenberg, J.B., Silverman, I. and Tambour, Y., 1998, *Combust Flame*, 113 (1–2): 271–273.
34. Gauthier, J.E.D. and Bardou, M.F., 1996, *J of the Institute of Energy*, 69 (479): 59–67.
35. Kang, Y.C. and Park, S.B., 1997, *Proc of ICLASS-1997*, 1122–1125.
36. Glover, A.R. and Bowen, P.J., 1991, Explosions in multiphase mixtures, ILIDS—Interferometric laser imaging for droplet sizing, *United Kingdom Explosion Liaison Group, December*.
37. Skippon, S.M., Natrass, S.R., Kitching, J.S., Hardiman, L. and Miller, H., 1996, *SAE International Paper 961204, International Spring Fuels & Lubricants Meeting, Dearborn, Michigan, 6–8 May*.

## ADDRESS

Correspondence concerning this paper should be addressed to Dr P. J. Bowen, Cardiff School of Engineering, Division of Mechanical Engineering and Energy Studies, University of Wales, Cardiff, Queens Buildings, The Parade, PO Bo 685, Cardiff CF2 3TA, UK. E-mail: bowenpj@cardiff.ac.uk

*The manuscript was received 17 July 1998 and accepted for publication after revision 23 November 1998.*