Fires, Explosions, and Venting in Nuclear Reactors

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ABSTRACT

A brief historical review covers salient reactor fires and explosions, principally centred around the use of graphite as a neutron moderator, and the high temperature generation of hydrogen in reactions of steam and zirconium. An alternative to uncontrolled, excessive, build-up of pressure, followed by uncontrolled explosion, is the provision of a buffer vessel, in which there is separation of hydrogen from radioactive products in permeable membrane separators. The hydrogen is then flared. Possible rates of production of hydrogen are compared, along with the rates at which it can be separated and flared in lifted jet flames, which give the highest burn rates. Cross winds can result in a transition to rim attached, downwash and wake-attached flames, all with a significantly reduced burn rate, or complete flame extinction. The performance of lifted jet flames of C_3H_8, CH_4 and C_2H_4, when exposed to increasing air cross winds velocities, are presented. These provide a basis for synthesising the performance of H_2 flames, also in cross flows. The H_2 relationship is rather different from that of the hydrocarbons, on account of the higher chemical reactivity of hydrogen, its small laminar flame thickness, reduced air requirement, higher acoustic velocity, and minimal flame lift-off distance. Destruction of hydrogen lifted jet flames by the cross flow of atmospheric air is significantly less likely than it is for propane jet flames. Flaring with micro-tubes might be advantageous for integrating flaring with membrane hydrogen separation, whilst high mass flow rates can be achieved with large diameter flares in the lifted flame, supersonic regime.

KEYWORDS: Hydrogen, jet flames, reactor venting, cross flow.

NOMENCLATURE

B	molar fuel/cross flow air rate ratio
C	molar fraction of air in combined molar flows of fuel and air into lift-off volume
C_c	critical value of C for reduction in U_b* by cross flow
C_p	constant pressure specific heat (J/kg·K)
C_{SL}	values of C, at the equivalence ratio for maximum laminar burning velocity, S_L
D	pipe diameter (m)
D_o	pipe external diameter (m)
f	ratio of fuel to air moles in fuel-air mixture for S_L
k	thermal conductivity (W/m·K)
L	flame lift-off distance (m)
u_j	mean fuel flow velocity at the exit plane of pipe for subsonic flow. For ratios of atmospheric pressure to P_i equal to, or less than the critical pressure ratio, or choked sonic velocity after isentropic expansion from P_i (m/s)
U*	dimensionless flow number for choked and unchoked flow, $(u_j/S_L)(\delta_k/D)^{0.4}(P_i/P_a)$
U_δ*	Value in Eq. (1) with $\delta = \nu/S_L$ in expression for U*

Greek

δ_k	laminar flame thickness, (m) $(k/C_p)_{To}/\rho_j S_L$
φ_{SL}	equivalence ratio for maximum laminar

Proceedings of the Ninth International Seminar on Fire and Explosion Hazards (ISFEH9), pp. 809-818
Edited by Snegirev A., Liu N.A., Tamanini F., Bradley D., Molkov V., and Chaumeix N.
Published by Saint-Petersburg Polytechnic University Press
ISBN: 978-5-7422-6498-9 DOI: 10.18720/spbpu/2/k19-9

$\overline{m}_c$ cross flow air mole density, (moles/m^3)
$\overline{m}_j$ fuel mole density, (moles/m^3)
P_a atmospheric pressure (Pa)
P_i initial stagnation pressure (Pa)
Re_c air cross flow Reynolds number, $u_c D_o / \nu_{air}$
S_L maximum laminar burning velocity of the fuel-air mixture in ambient atmosphere (m/s)
To temperature at inner layer of laminar flame (K), see [17]
u_c cross wind velocity (m/s)
burning velocity
ν kinematic viscosity, under conditions of ambient atmosphere (m^2/s)
ρ density (kg/m^3)

Subscripts

a ambient conditions
air air
b value at blow-off
c cross flow air
i initial stagnation conditions
j jet fuel

INTRODUCTION

Four key reactor combustion incidents are briefly summarised in Table 1. This is followed by more detailed considerations, which identify key problems, particularly the rapid formation rate of undesirable products at high temperature, their inadequate containment, and the feasibility of venting at an adequate rate to the atmosphere.

Table 1. Some key nuclear reactor fires and explosions

Incident	Problem	Remedy
1957 Windscale. Plutonium production. Graphite moderated, air cooling, graphite fire.	Temp. > 380 °C during Wigner release. Metal melted. Fire increased by increased air flow.	Fire further increased by CO_2. Water risked H_2 generation, but “If it goes up we go with it”.
1979 Three Mile Island. Pressurised Water Reactors (PWR), loss of coolant.	Core melted. Zircalloy and steam generated H_2 bubbles at top of reactor.	Hydrogen bubbles, at top released in stages, then mixed with air.
1986 Chernobyl. Boiling Water Reactors (BWR). Graphite moderator cooling by He/N_2. Uncontrolled rising temperature. Steam bubble increases reactivity.	Zirconium/steam generated H_2. Steam explosion probably followed by hydrogen explosion.	Excessive overpressure destroyed reactor. Extensive fires combatted with water.
2011 Fukushima. BWR. Coolant failure due to earthquake and tsunami.	Hot zirconium cladding reacted with steam after water level dropped, producing H_2. 3 reactors had meltdowns and 1 building destroyed.	All 4 reactors had H_2 explosions. Injection of water, including seawater. Concerns about H_2 explosions.

GENERAL CONSIDERATIONS

The 1957 Windscale fire arose from the use of graphite to moderate neutron energies in the course of plutonium production. Associated Calder Hall power reactors used CO2 as a coolant which, at high temperatures could react with the graphite. The Windscale fire was caused by overheating of the coolant air above 380 °C during Wigner energy release. This caused the graphite to burn in the coolant air. During the fire CO_2, was used as an attempted coolant, but it increased the combustion rate. At some risk of excessive H_2 generation, H_2O was successfully employed [1]. Graphite moderation was employed at Chernobyl and in the subsequent fire, after loss of control of the reactors, the graphite became incandescent, with the formation of CO. This burned, along with the fuel cladding. Additional difficulties are created in reactors by neutron-induced material degradation [2]. A problem with graphite in such reactors as the Advanced Gas Cooled Reactor, is the neutron displacement damage to the graphite structure. It is difficult to replace the damaged graphite, during the reactor lifetime, although this was achieved with the St. Petersburg reactor. Containment prior to the Chernobyl incident was inadequate.

At Three Mile Island, through loss of coolant, about half the reactor core melted. Radio-nuclides remained inside the reactor or dissolved in water. Reaction of steam with the zirconium cladding of the fuel rods generated H_2. Corrosion rates of zirconium become 10 times greater inside a reactor [2]. There was no major breach of the containment. Power failure prevented venting of the primary containment up the 100 m high stack. Some of the gases seeped into reactor buildings, accidently exploded, and removed parts of the structure. The widespread dispersal of H_2 would weaken the mixture, reducing its laminar burning velocity and whilst, initially, turbulence would enhance the burn rate, ultimately it could extinguish it. It has been estimated that a rather lean, near-homogeneous mixture of 8% H_2/air burned, creating an overpressure of 190 kPa [3], and reducing the overall damage. Figure 1, from research sponsored by the UK Atomic Energy Authority at the time, shows the development of flame quenching with increasing turbulence in weak mixtures of 6, 8, and 10% hydrogen (stoichiometric % = 29.6) [4].

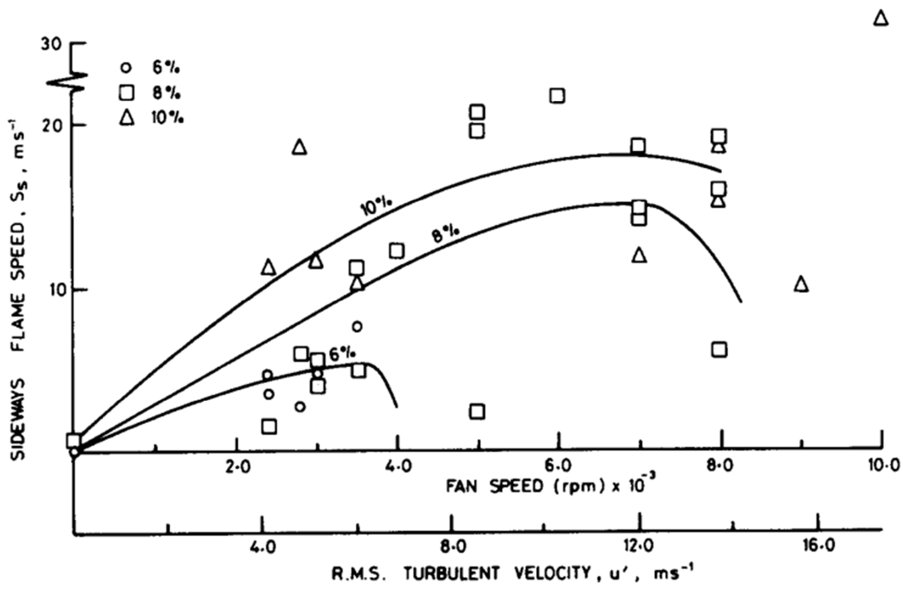

Fig. 1. Quenching effect of the rms turbulent velocity on hydrogen-air flame speed, taken from [4].

After this incident, reactor owners were required to strengthen venting systems to prevent leakage of H_2 into secondary containment buildings. Most of the H_2 was generated from Zircalloy cladding reacting with steam. It has been estimated that 1,200 kg of H_2 would be created, were all the cladding to be be oxidised by steam, and that complete combustion of the zirconium in a 1,000 MW(e) reactor would release $198 \cdot 10^9$ Joules [2].

Hydrogen and O_2 can also be formed in light water cooled nuclear reactors by the radiolytic decomposition of water [5]. If significant amounts of H_2 and O_2 were to be created by radiolysis in stoichiometric proportions, this would be very serious because of the very high reactivity of such a mixture [6]. However, Gordon et al. [7] found this not to be so, with no more than 0.7% H_2 created by radiolysis, and this was removed by recombination. To avoid explosive recombination with O_2, many reactors have been retrofitted with passive hydrogen recombiners within the containment. One approach, implemented in a few Boiling Water Reactors, has been to burn the H_2 inside the containment using distributed glow plugs [8]. Other remedial action has involved injection of N_2 into the reactor. At Fukushima, reactors survived the earthquake, less so the tsunami. There was no reactor cooling an hour after shut down. At both Three Mile Island and Fukushima there was a failure to remove the radioactive decay heat from the fuel [9].

At Fukushima all the fuel in Unit 1 melted, much of it leaking out. Seawater with neutron absorbing boron were used as coolant, but reactors overheated for many days. The reactors were GE/Toshiba/ Hitachi Boiling Water Reactors, operational since 1971-75, with powers ranging from 460 to 1,784 MW(e). Pressure built up in Units 1 to 3, with most of the fuel melting [9]. Venting was designed to be through an external stack, but, in the absence of power, most of it back-flowed into the top floor of the reactor building. Venting began almost 24 hours into the emergency [10]. Containments were vented to atmosphere. Hydrogen leaked into reactor buildings and caused large explosions in Units 1, 3 and 4. Each Unit is estimated to have produced 800-1,000 kg of H_2. Hydrogen explosions caused tremendous damage. Even when fissioning had ceased, significant heat was generated through radioactive decay.

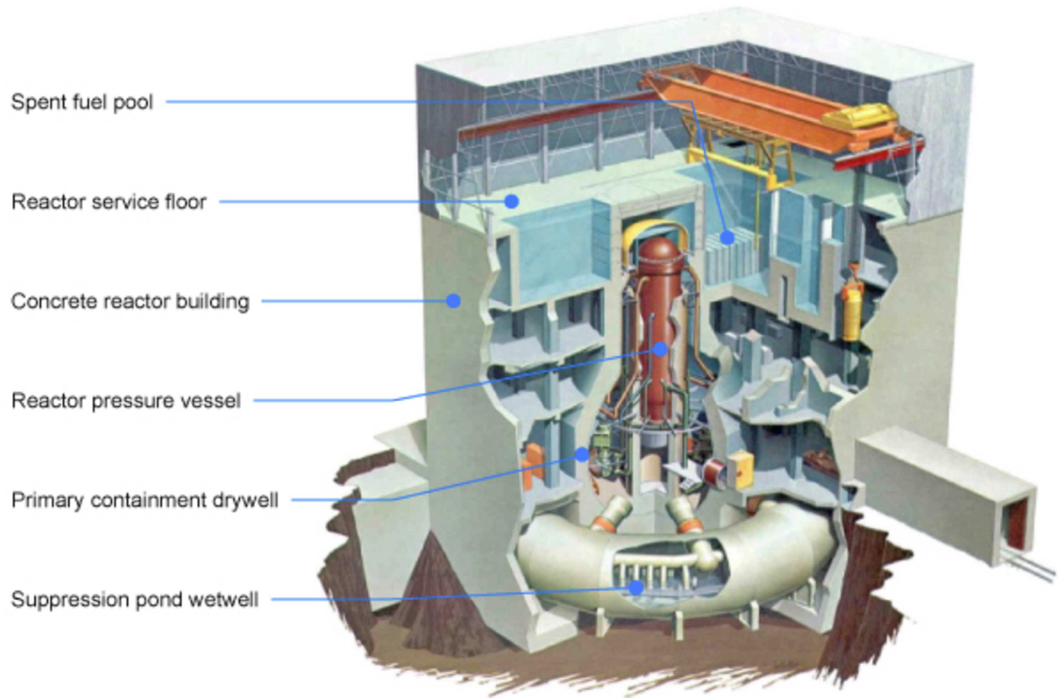

Fig. 2. Containment of Reactor Pressure Vessel, taken from [9].

As a consequence, the three Fukushima reactor cores, see Fig. 2, melted in the first two or three days of the emergency. There were considerable releases of radio nuclides and cooling water, with a total of ten core melts. The rate of formation of H_2 was controlled by the rate of oxidation of the zirconium fuel cladding by steam, at about 1,300 °C [5]. This rate of reaction was far beyond the capability of H_2 recombiners, N_2 inerting, and the time required to ensure the requisite purity of vented gases. This poses the current major challenge.

THE CHALLENGE

In a loss of coolant, or similar crisis, the reactor and its immediate containment are of inadequate volume to contain all the hydrogen that might be produced, as is evident from the relatively small Primary Reactor Containment shown in Fig. 2. Boiling Water Reactors operate at pressures of about 8 MPa, while Pressurised Water Reactors, with a secondary circuit, operate at about 16 MPa. Were venting to be long delayed a worse situation would arise from failure of the reactor/containment. Unless it is well controlled, allowing emergency venting to atmosphere too early will disperse undesirable radio-nuclides and increase the probability of uncontrolled hydrogen explosions. This perspective leads to the necessity of a large buffer vessel into which the primary products are vented. Ultimately, large amounts of H_2 must be vented, preferably free of undesirable radio-active products. This might be achieved by separating and containing such products, while the hydrogen would be contained and flared in a controlled manner.

Hydrogen separation has been proposed, through the use of hydrogen gas permeable membrane separators in a stream rich in H_2. This could be passed through a charcoal adsorber to adsorb radio-active particles and then flared in a gas burner [11, 12]. Inability to control the build-up of the high temperature reaction products, inadequate venting rates, particularly of hydrogen, and crisis management have been characteristic features of the described malfunctions. An essential requirement is a large buffering volume to contain the products during their initial high rates of formation. It is also desirable to separate and contain the most damaging products, whilst flaring hydrogen as soon as possible, in order to prevent its build up. A safe balance must be sought between rates of H_2 production, separation, and flaring. Although flaring of H_2 is not essential, if the release is large, it is a safeguard against its hazardous accumulation elsewhere. The hydrogen flaring process is now briefly considered, in terms of its feasibility for achieving adequate burn rates, the practicality of flaring, and the ability of flares to withstand cross winds.

CONTROL OF HYDROGEN FLARING

Limitations due to blow-off

Jet flames exhibit a variety of structures, ranging from lifted flames with high burn rates, in which the fuel jet flow is dominant, to rim and turbulent wake flames, the latter stabilised by the wake of a strong air flow across the fuel pipe. Flame blow-off and extinctions in the former case occur at sufficiently high values of the flow number, U^*. In the latter, they occur when the Reynolds numbers Re_c, based upon the pipe outer diameter and cross flow air velocity becomes sufficiently high [13]. Between these limits are a variety of other structures ranging from rim-stabilised to downwash flames. Because of the importance of their higher burn rate, lifted flames will be considered in detail, together with the effects of an increasing cross flow of air. The parameter U_b^* was formulated on the bases of both stretched laminar flamelet mathematical modelling [14] and the experimental derivation, correlation, and validation of appropriate dimensionless groups. Data were drawn from a vast experimental data bank [15]. This covered jet velocities, burning velocities, emitting plume heights, flame lift-off distances, and flame heights, involving six different fuels.

Flaring consists of the burning of a jet of excess fuel in the atmosphere. The highest burn rate within the reaction zone is achieved at the leading edge if the lifted flame, with flamelets burning at the maximum laminar burning velocity, S_L. The lift-off distance, L, is the distance between the exit plane of the pipe and this leading edge. If the ratio of fuel pipe diameter, D, to laminar flame thickness, δ_k, [14] is too small, there is difficulty in maintaining combustion, and the flame is soon quenched by excessive air entrainment.

Figure 3 shows experimentally-based correlations of the dimensionless flow number, U_b*, at blow-off, for different values of δ_k/D_b [16]. There is no cross flow and data are shown for four fuels. The flow number on the x axis, is defined as U_b*, = $(u_j/S_L)(\delta_k/D_b)^{0.4}(P_i/P_a)$, where u_j is the mean fuel exit velocity, D_b is the pipe diameter for blow-off, and (P_i/P_a) is the ratio of upstream stagnation to atmospheric pressure. Uniquely, in H_2 flames, the high diffusivity of H atoms induces significant heat release earlier in the flame [17]. This necessitates a different approach in the use of flame thickness in generalised correlations [18]. This thickness is given by $\delta_k = (k/C_p)_{To}/\rho_j S_L$ [18] for the leading maximum laminar burning velocity. The data in Fig. 3 are experimentally based, from [16], except for C_2H_4 from [19], and are overwhelmingly from the subsonic pre-choked regime.

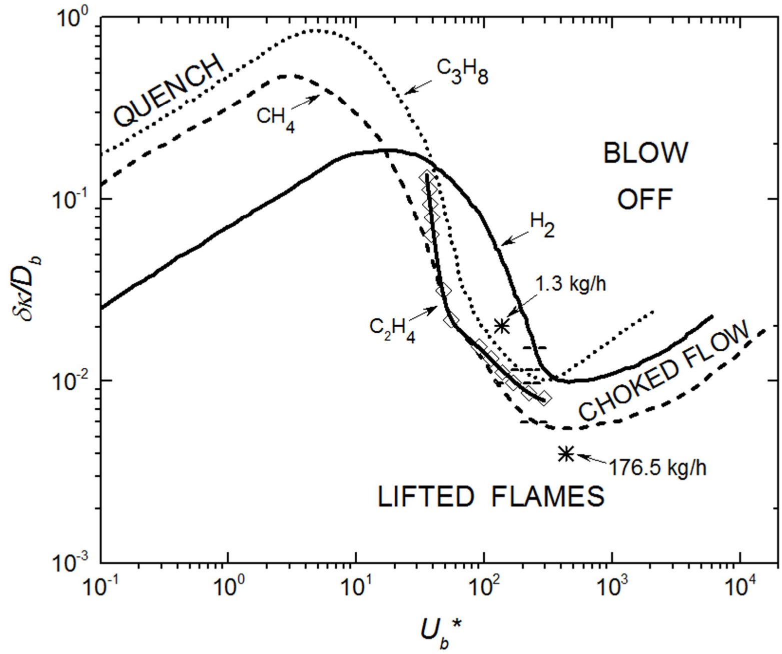

Fig. 3. Sonic and subsonic lifted jet flame blow-off and quench boundaries, for C_3H_8, CH_4, C_2H_4, and H_2. Short dashed horizontal lines show critical pressure ratio condition.

Locations at which the critical pressure ratio is attained on each blow-off curve in Fig. 3 were found from the compatibility of δ_k/D_b and U_b* at this pressure ratio. These are indicated by the short horizontal broken lines, below which flow is choked at blow-off. Below the blow-off curve, U_b*, for a given fuel, towards the lower values of δ_k/D_b, is the regime of lifted flames, with larger pipe diameters, above which stable lifted flames can be maintained. Above the curve, is the regime of decreasing pipe diameters, below which blow-off occurs.

Hydrogen flaring, with S_L = 3.03 m/s, is analysed, in terms of these generalised characteristics of lifted jet flames. First, in Fig. 3 the use of a micro-tube, D = 2.0 mm, δ_k = 0.03985 mm [16, 18], δ_k/D_b = 0.02, is considered. The initial/atmospheric pressure ratio is P_i/P_a = 1.8, just within the subsonic regime, before choked flow develops. Because of the high acoustic velocity of H_2, arising from the low molecular mass, the exit velocity, u_j, is also high, at 1,159 m/s, and U_b* = 144. These conditions give a micro-tube mass flow rate of H_2 of 1.3 kg/hour, indicated by the upper asterisk in Fig. 3.

Now consider blow-off in the choked flow regime, with P_i/P_a increased to 10 and D = 10 mm. In this regime, the reaction rate is enhanced by shocks and supersonic flows, at high U_b*. Now δ_k/D_b = 0.004, and u_j is equal to the acoustic velocity of 1,202 m/s, with an associated density of 0.51924 kg/m^3. These conditions yield U_b* = 436, a mass flow rate of 176.5 kg/hour, indicated by the lower asterisk, and a jet flame heat release rate of 6.9 MW. The generalised data in [15] suggest the jet flame height would be 4.8 m.

Hydrogen has a number of characteristics contributing to high jet velocity flames: a high laminar burning velocity, small flame thickness, small air requirement, and a high acoustic velocity. In contrast, its high reactivity makes it more prone to flame flashback from premixed flames.

LIMITATIONS DUE TO AIR CROSS FLOW

A further desirable characteristic in the flared venting of jet flames is an ability to survive the cross winds that might occur in the atmosphere. Available experimental data, on the effect of cross wind on U_b* have been re-expressed, but for pre-choked flow only, in terms of a parameter *C*. This is the mole fraction of cross flow air in the mixture that is created with the jet fuel, within the lift off distance, *L*. This has a volume $(\pi D^2/4)L$, with measured steady fuel jet and air cross flows into it. The resulting experimental data for U_b*, processed in this way for C_3H_8, and CH_4, from [19], are plotted against values of *C* for different D/δ_k, in Fig. 4. Stable, smaller diameter, jet flames exist within the peninsula. At the upper, lifted flame, limit, blow-off occurs, and at the lower limit slower burning flames are no longer lifted, but are attached to the burner.

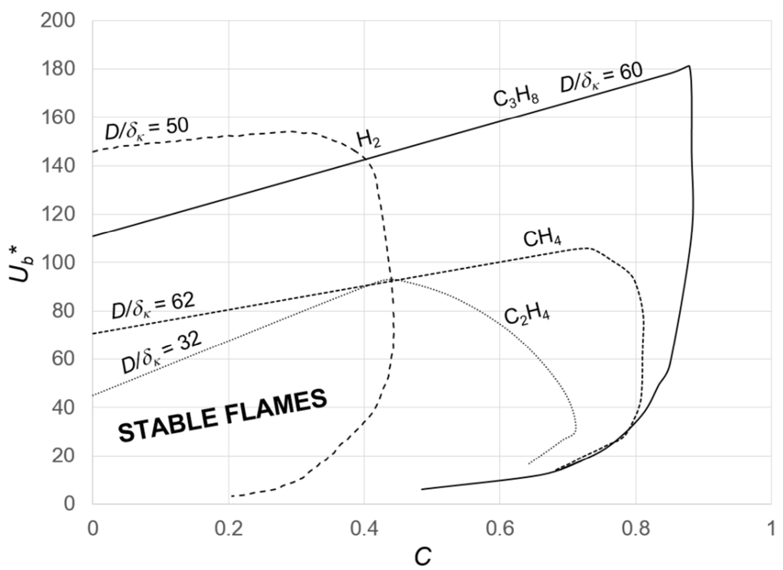

Fig. 4. Blow-off limits of stable CH_4, C_2H_4 and C_3H_8, lifted flames with air cross flows, in terms of U_b* versus *C*, for different values of D/δ_k. The dashed curve for H_2 is synthesised from the data of the other fuels.

Initially, as the air cross flow velocity, u_c, and *C* increase from zero, the mixture within the lift-off volume becomes more reactive. This causes the blow-off velocity and U_b* to increase. Other similar stability peninsulas for these gases, at different values of D/δ_k, can be constructed from the data in [19]. The experiments show that eventually increasing the cross flow necessitated significant reductions in the fuel flow rates and the values of U_b* that could be sustained, at a critical value of *C*, = C_c. This occurs before *C* has attained a value, C_{SL}, = $(1 + \varphi_{SL})^{-1}$, at which the associated equivalence ratio in the lift-off volume is that for S_L, namely, φ_{SL}. Both of these values of *C* are given in Table 2 for the different fuels. Of course, additional air is still entrained by the jet downstream beyond the lift-off distance. At C_c, the flow number U_b* must decrease in order to sustain the flame. Ultimately there is a transition to the lower limb of the peninsula and u_c is much reduced. For smaller values of D/δ_k, values of C_c, become less clearly defined. An example of this is indicated by the dotted curve for C_2H_4 in Fig. 4.

For the C_3H_8 peninsula in Fig. 4, at C_c the experimental data show quenching to begin at u_j = 242 m/s, with u_c = 5 m/s, as U_b* begins to fall sharply. For the CH_4 peninsula comparable values are u_j = 192 m/s and u_c = 3 m/s. These low values of u_c suggest that quite moderate cross flows can jeopardise the stability of lifted flames.

No cross wind data could be found for H_2, but from the experimental cross wind data available for the other three gases, it was possible to estimate the performance of hydrogen lifted jet flames in air cross flow. Another important factor controlling lifted jet flames in the presence of cross flows, is the ratio, f, of fuel to air moles at φ_{SL}. Values of f also are given in Table 2. Hydrogen requires significantly less air than hydrocarbons, and the value of f for H_2 is more than ten times higher than that for C_3H_8. As can be seen from Table 2, its C_{SL} value of 0.569, is significantly lower than those for the hydrocarbons. It was found that, for the same value of D/δ_k, C_c tends to decrease with C_{SL}. Consequently, the value of C_c also will be low. Guidance about the extent of this deficit below C_{SL} for H_2 was obtained from consideration of that occurring for other gases. This led to the tentative assignment of a value of C_c = 0.44 for H_2, with a value of U_b* of 144 at C = 0, and D/δ_k = 50, taken from Fig. 3. These considerations enabled the tentative, dashed, characteristic peninsula for H_2, shown in Fig. 4, to be constructed.

From the material balance of jet and cross flows in the lift-off volume, it follows that that $C = 1/(B + 1)$. Here $B = (u_j\overline{m}_j/u_c\overline{m}_c)\pi/4(D/L)$, in which the first bracketed term is the ratio of fuel to air molar fluxes into the lift-off volume. Evaluation of u_j/u_c requires the normalised lift-off distance for H_2 lifted flames, and this was found from the expression given for L/D, based on a different expression for flame thickness in U_δ* [24]:

$$(L/D)f = -0.0002U_\delta^{*2} + 0.19U_\delta^* - 3.3. \tag{1}$$

This remains valid as the cross flow develops, but it becomes increasingly unreliable beyond C_c. The expression for B yields values of u_j/u_c, at the onset of the rapid decline in U_b*. The value of U_b* then gives that of u_j.

Table 2. Property values and references for characterising Fig. 4

Fuel	D/δ_k	Ref. S_L	φ_{SL}	S_L, m/s	f	C_{SL}	C_c
C_3H_8	60	[20]	1.1	0.43	0.046	0.956	0.93
C_2H_4	32	[21]	1.2	0.72	0.084	0.923	0.65
CH_4	62	[22]	1.02	0.39	0.107	0.903	0.81
H_2	50	[23]	1.8	3.03	0.756	0.569	(0.44)

These various considerations suggest that for H_2, with U_b* = 144 and u_j = 1159 m/s, with C_c = 0.44, D/δ_k = 50, and f = 0.756, then u_c = 62 m/s. A natural atmospheric cross wind as high as this is uncommon. It is therefore unlikely that a lifted, venting hydrogen flame could be significantly disturbed by atmospheric conditions, and make the transition to a slower burning attached flame. This behaviour contrasts with that for C_3H_8 and CH_4. The high value of f for hydrogen reduces the air requirement and lift-off distance, while the high acoustic velocity is associated with high jet velocities at a given Mach number. These can be subsonic and in excess of 1,000 m/s.

CONCLUSIONS

1. Loss of coolant and other malfunctions can result in reactors over-heating and creating a variety of chemical reactions and heat releases. This must also be viewed in the context of improving operational efficiencies by operating reactors at higher temperatures. The least acceptable consequence of these is uncontrolled reactor failures, with the release of radioactive products and explosive gases into the atmosphere. Ideally, such a release could be avoided by early venting of the reactor, without any release of noxious products and flammable gases, and no external explosion. This might be achieved by venting the reactor into a much larger buffer vessel, in which the hydrogen might be wholly or partially separated and then flared.

2. In normal operation, H_2 recombiners can process about 195 kg/h of H_2, but in the case of an accident, the required rate would increase 100 to 400 fold [11], beyond the capabilities of this technique. In this situation, it has been proposed that, after removal of the water from the gaseous mixture, the H_2 should be separated, using a gas permeable membrane separator [11]. The H_2 stream would then pass through a charcoal adsorber to remove radioactive products, before being finally flared. If choked flow flaring on a 10 mm pipe, as demonstrated in Fig. 3 at a rate of 176.5 kg/h, were to be employed, the accumulated approximate estimate of 3,000 kg of H_2 at Fukushima would be flared on three such burners in just under 6 hours. There are many ways in which H_2 membrane separation can be implemented, covering a rich variety of materials, and structures [25].

3. With hydrogen permeable membrane separation, it is suggested in [11] that the differential pressure across the membranes should not exceed 1.724 MPa. If the H_2 were to be stored at 2 MPa and 300 K, 1,000 kg of H_2 would occupy a volume of 1,638 m^3, a cube with a 11.8 m side. This is a practically convenient size, which might combine storage and separation. The present analyses of the subsequent flaring have shown that the characteristics of H_2 are particularly well suited to a flexible approach to storage, separation and flaring, albeit with some possible delay for H_2 separation if dispersal of harmful radio-active products is to be avoided.

4. Flaring of H_2 is favoured by its low air requirements which leads to compact lifted flames. Its high acoustic velocities, arising from its low molecular mass, combined with its high burning velocity, lead to high values of fuel jet velocity. Although the analysis of air cross flow on H_2 lifted flames provides only an estimate of velocities, rather than accurate predictions, it nevertheless clearly shows that the extinction of lifted flames due to atmospheric cross winds is unlikely. The same cannot be said of C_3H_8 and CH_4 flames. There is clearly a need for experimental data on H_2 lifted jet flames in cross flows.

5. A unique aspect of H_2 jet flames is their ability to support micro-jet flames, a consequence of their low δ_k values. This could be relevant also in the separation process. Another possibility is for vented gas from the reactor to be immediately flared in micro-jets, followed by removal of radio-nuclides.

ACKNOWLEDGEMENTS

A.P. gratefully acknowledges the financial support of the Royal Society in the form of a Postdoctoral Newton International Fellowship.

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