

FRAGMENTATION IN THERMAL EXPLOSIONS

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(Received 29 March 1973 and in revised form 6 August 1973)

Abstract—A series of experiments involving explosions between molten tin and water is described. The results, together with information from other work, indicate that thermal explosions usually involve several distinct interactions in the same mass of material, and that each interaction forms a bubble containing some vapour whose collapse initiates the next interaction. It is thus shown that a small disturbance can escalate by successive growth and collapse cycles into a large explosion.

It is postulated that vapour collapse is the main cause of dispersion in many thermal explosions. A possible mechanism for this is that during collapse a high-speed liquid jet is formed (as observed in cavitation damage studies), which penetrates and disperses in the molten material. It is shown that the jet penetration hypothesis can account for both the timescales and energy transfer rates characteristic of thermal explosions, including those in shock tube geometry.

NOMENCLATURE

- P , pressure;
- r , radius;
- T , lifetime of bubble;
- V , volume;
- σ , surface tension.

Subscripts

- v , vapour;
- ∞ , ambient.

1. INTRODUCTION

THE CONTACT of two liquids, one at a temperature significantly above the boiling point of the other, can lead to explosive vapour formation and cause mechanical damage. Such events can present a safety problem and are also of economic importance in the nuclear, smelting, and liquid natural gas industries.

Experimental investigation has shown that thermal explosions on a laboratory scale occur in times of 10^{-3} – 10^{-4} s, producing pressures between 10^2 and 10^3 bars. It is now generally accepted that such pressures can only be explained in terms of known heat transfer rates if there is significant enhancement of the interfacial surface area [1–3]. The mechanical energy required to produce this rapid area enhancement against liquid inertia and interfacial tension ($\sim 10^3$ erg cm^{-2}) must in general derive from the thermal energy, but it is not yet resolved whether the

enhancement occurs in a separate single-stage initiating process, or whether the explosion is itself capable of generating its own surface area. The logical sequence of explosion processes is summarized in Fig. 1.

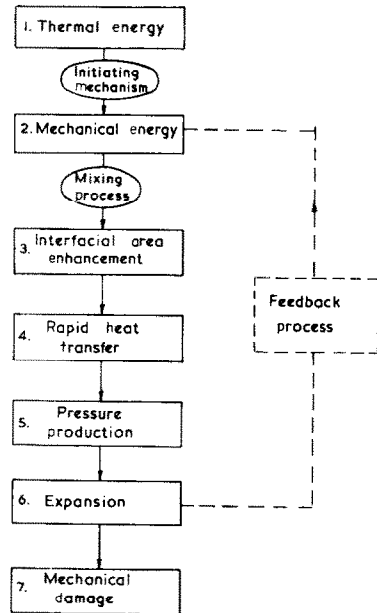


FIG. 1. Chain of processes occurring in a thermal explosion.

Much of the theoretical work to date [4–6] has been concerned with the problem of calculating the pressures and explosive yields resulting from heat transfer to the coolant (i.e. stages 4–6 in Fig. 1). Most models assume thermodynamic equilibrium, but some progress has been made towards specifying realistic energy distributions and heat transfer rates when phase changes or rapid variations of coolant properties with temperature are included [7, 8]. However, the remaining area of uncertainty concerning the extent and speed of the area enhancement process is most important, since the overall energy transfer rate depends directly on the magnitude of the interfacial area.

Experimental work on area enhancement (i.e. mechanical mixing of two fluids) has led to the identification of several single-stage initiating processes which could act to convert some of the initial thermal energy into mechanical energy [9–11]. Mechanisms so far suggested rely either on violent (or transition) boiling, or on boiling of trapped coolant.

(a) *Transition boiling* of the coolant is most likely to occur when part of the hot surface has cooled sufficiently that the film boiling vapour blanket becomes unstable and intermittent wetting occurs [12]. A current hypothesis for LNG explosions [13] is that liquid/liquid contact occurs at this stage, and because of this nucleation of the coolant is suppressed until the coolant superheat reaches some limiting value (the homogeneous nucleation temperature). Thus the onset of transition-boiling is particularly violent with potential mechanical yields $\sim 2 \text{ Jcm}^{-2}$ [14]. It is suggested that this is sufficient to account directly for the full yield of the explosion, but it is difficult to understand on this basis how relatively intense explosions can occur. The theory has also been applied to sodium and UO_2 , [15] though in this case contact and freezing must at best be simultaneous, but more fundamental problems arise in translating the hypothesis to molten aluminium and water, where the instantaneous contact temperature is above the critical temperature of the coolant so that wetting and superheat are not meaningful concepts.

Normal transition boiling will still occur when the surface has cooled sufficiently (and solidified), and it seems likely that this process could be energetic enough to cause rapid mixing if adjacent material is still molten. The mixing may occur either directly from the nucleation pressures, or indirectly via subsequence vapour collapse, though the mechanical energy available in the collapsing blanket ($\sim 10^6 \text{ erg cm}^{-3}$) would be only sufficient to produce the necessary dispersion if the blanket volume reached a significant

fraction of the volume of the molten material; such a thick blanket should be easily visible in a high speed film record, but has not been observed in the experiments to date.

(b) *Liquid entrapment* is most likely to produce rapid dispersion if the coolant is trapped in a frozen shell which is burst by coolant vaporisation. There is evidence that such shells occur (the outside of a drop must of course freeze first) and that entrapment within them is possible for a limited range of Weber numbers [16, 17], but it is difficult on this hypothesis to explain explosions where no freezing is possible (e.g. hot mercury in water). The alternative possibility is that violent boiling will be engendered in a small quantity of coolant solely by virtue of its entrapment. This seems unlikely unless there has been significant prior dispersion such that the entrapped drops are very small (i.e. $< 10^{-3} \text{ cm}$).

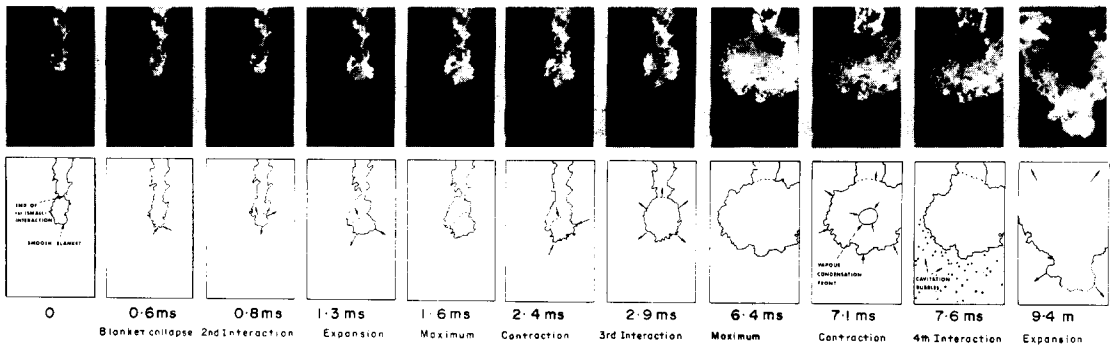
It may be necessary to invoke both transition boiling and entrapment, and perhaps other mechanisms also, to explain the triggering of explosions in a wide range of materials and conditions. However, since neither mechanism leads directly to predictions of the speed and degree of dispersion, it is not clear whether either is sufficiently effective to act as a single-stage area enhancement process. If not, then a feedback process (as shown in Fig. 1) whereby the explosive energy transfer can itself cause dispersion is required. This seems more likely to produce a genuinely explosive interaction, but as yet no convincing physical mechanisms for the feedback process have been postulated.

It is the purpose of the present paper to describe an experimental investigation into the causes of dispersion and, with the aid of the data obtained and information already available from other work, to suggest a physical mechanism for the feedback process which may be responsible for causing most of the dispersion in a wide range of thermal explosions.

2. EXPERIMENT

In the present work three types of experiment have been performed using molten tin in water. In each experiment, the interaction was filmed at 8000 ps^{-1} and pressure records obtained from an immersed transducer with a flat frequency response up to 200 kHz.

(a) In the first type of experiment, a few grams of tin were dropped from 2 to 5 cm above an open tank of water about 30 cm deep. It was found that explosions occurred only if both the tin was above 400°C and the water was below 60°C , confirming the thresholds reported previously [10, 18]. Frames



Sequence from a film showing development of a thermal explosion (T_{in} at 800°C ; water at 20°C)

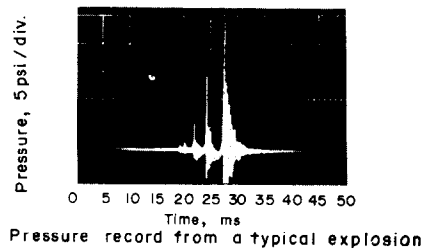


FIG. 2. Photographic and pressure records of molten tin/water thermal explosion.

from a film of a typical explosion in water at 20°C with the tin initially at 800°C are shown in Fig. 2. There are four interactions of increasing violence occurring in the same drop of tin. In each case the approximately spherical bubble-like region of two-phase coolant and debris grows and then contracts slightly before the next interaction. In the period following the third interaction, a brighter vapour-rich region is visible within the 'bubble' which moves inwards and collapses completely during this period. The subsequent interaction is initiated at the moment of vapour collapse. In the last and most violent interaction, the occurrence of the interaction pressure pulse can be inferred from the sudden appearance, immediately before final, explosive expansion, of small bubbles in the bulk of the water; these are believed to be cavitation bubbles produced by the passage of the rarefaction wave occurring when the pressure pulse is reflected from the free surface. Pressure records for other tin dropping experiments confirmed the occurrence of multiple interactions (see Fig. 2); between pressure pulses are periods during which the pressure remains a little below ambient,

as would be associated with the decelerating growth and accelerating collapse of a vapour bubble. The duration of each low-pressure period corresponds approximately to the expected lifetime of a bubble of the observed radius ($T \sim 1\text{--}10$ ms for $r \sim 0.5\text{--}5$ cm).

(b) It occurred to us that the existence of a water temperature (or subcooling) threshold for explosions could be utilised in an experiment to produce explosions at will in an immersed stationary drop. In the apparatus shown in Fig. 3, molten tin was poured on to a crucible beneath the surface of the water which was maintained at low pressure (~ 0.1 bar) and at a temperature ($\sim 40^{\circ}\text{C}$) which, for that pressure, was above the subcooling threshold.

The pressure was then suddenly increased to 1 atm. Multiple explosions were observed occurring reproducibly, starting spontaneously during the pressure rise time (20 ms).

A typical film of the behaviour of the drop (Fig. 4) shows saturated film boiling changing to subcooled film boiling as the pressure rises. It may be seen clearly on the original film that a small interaction occurs between the base of the drop and the crucible, followed

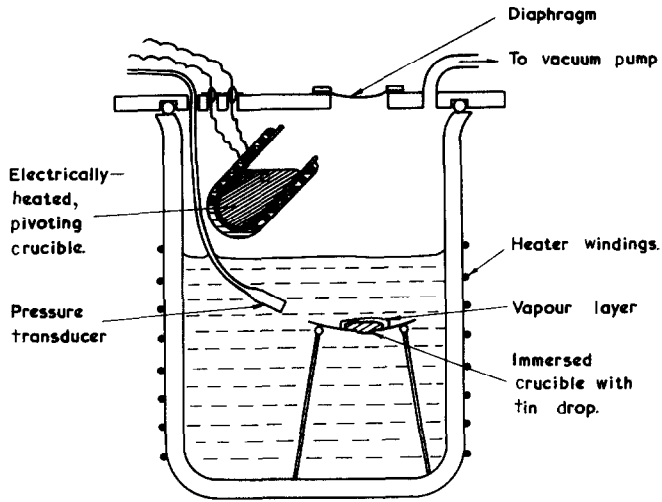


FIG. 3. Apparatus for testing the effect of a sudden pressure change on a stationary tin drop.

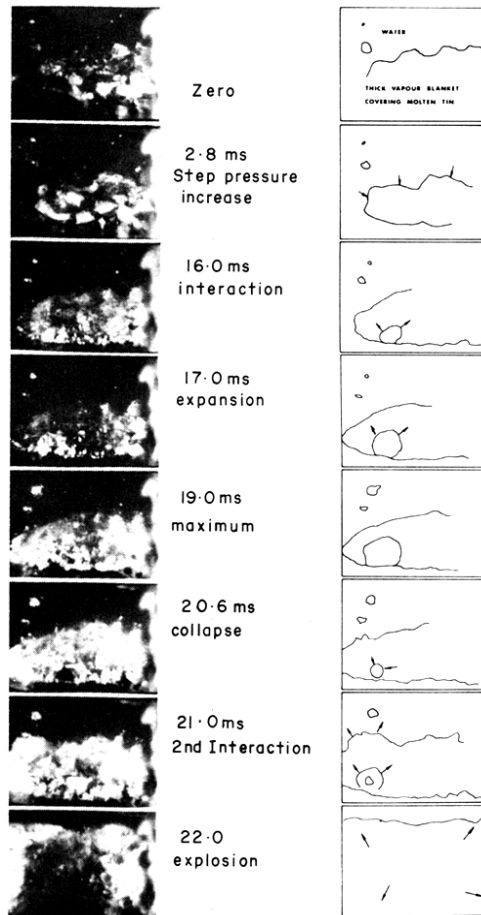
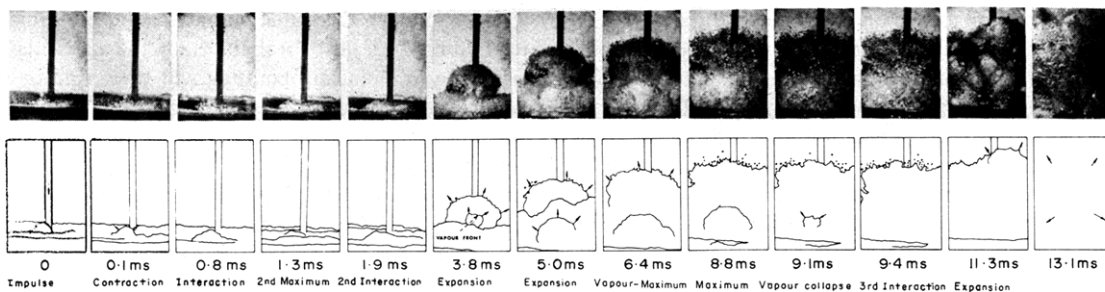


FIG. 4. Molten tin/water thermal explosion initiated by step increase in ambient pressure.

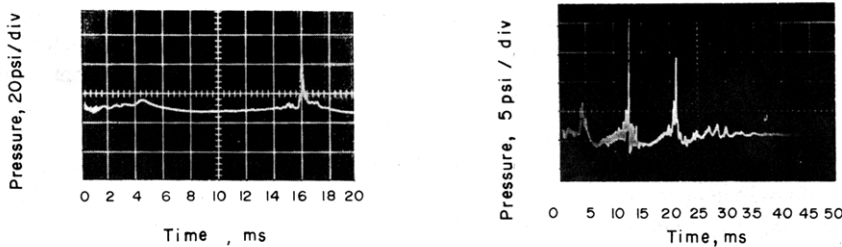
by a second, larger interaction producing a bubble 3 mm radius. This bubble collapses and a violent explosion occurs, centred at the location of the bubble. During the expansion a number of minor eruptions occur from the surface of the drop indicating that other explosion centres not visible in the film are probably operating almost simultaneously with the one observed.

hammer blow) transmitted via a steel rod to the loosely-mounted crucible. It was shown by a separate experiment with a similarly-sized drop of Bi-Pb-Sn-Cd alloy (melting point 70°C, and hence molten at the water temperature) that the impulse was not sufficient per se to cause any dispersion.

It was found that the impulse triggered multiple thermal interactions in the molten tin drop. Typical



Sequence from a film showing development of an impulse-initiated thermal explosion (tin at 800°C: water at 80°C)



Pressure records from typical impulse-initiated explosions

FIG. 5. Photographic and pressure records of impulse-initiated molten tin/water thermal explosion.

Pressure traces from these experiments again showed multiple peaks with periods of low pressure associated with bubble motion. Peak pressures of between 2 and 4 bars were recorded about 6 cm from the explosion centre. The interaction region pressure clearly depends on the assumed radius of the region at the time of measurement, but by taking a somewhat arbitrary value of 3 mm, interaction pressures of 30–60 bars are obtained.

(c) In the third type of experiment, a mechanical disturbance was applied to the otherwise stable situation of a tin drop supported on a crucible under water at 80°C (i.e. above the subcooling threshold). The disturbance was in the form of an impulse (a light

pressure traces initiated by the impulse (Fig. 5) showed that the first interaction occurred about 5 ms after the impulse, and was followed by a lower pressure region, characteristic of bubble growth and collapse, lasting about 10 ms, followed in turn by a second interaction peak. A third and sometimes a fourth interaction followed at similar intervals but less reproducibly. The film records (Fig. 5) show an event in which the impulse moves the crucible slightly downwards, causing the vapour blanket surrounding the tin drop first to grow (by a few millimetres) and then to collapse. The blanket collapse initiates a minor interaction, which produces a small vapour bubble which grows and collapses. This collapse

initiates a more substantial interaction, which produces a rapidly expanding hemispherical interaction region. Small bubbles existing in the water have been seen to be engulfed by the growing interaction region, showing the liquid-phase content of the bubble to be increasing. The growth then slows and a bright vapour region, as seen in the spontaneous explosion film (Fig. 2), moves in and collapses completely at the moment of the third interaction.

The experiment was repeated at 95°C at which temperature vapour would condense only very slowly, and no explosion could be produced.

Experiments at 80°C in which some dispersal was produced mechanically (by driving a 2 cm dia piston through the drop at $\sim 20 \text{ cm s}^{-1}$) generally resulted in a weak single interaction delayed significantly from the dispersal impact. Thus pre-dispersal appears to prevent multiple interactions, possibly by removing the molten material from the site of bubble collapse.

3. DISCUSSION

These experiments suggest that thermal explosions commonly involve the occurrence of several distinct interactions in the same mass of material, and that each interaction produces a vapour region which, on collapse, triggers the next interaction. A small interaction may in some circumstances escalate in this manner in several growth and collapse cycles into a violent explosion. We interpret this as showing that vapour growth and collapse can operate as a feedback mechanism whereby explosive energy release is returned to the interaction region as mechanical energy which can create more dispersion and thus produce a larger explosion.

The mechanism initiating the first interaction was visible only in the case of the mechanically perturbed system when that interaction was of significant size. Under these conditions the initiation could be directly attributed to the collapse of the thick vapour blanket surrounding the drop.

In the case of the experiment with a free-falling drop, there is indirect evidence from the explosion threshold water temperature that the initiation of the first interaction is due to the onset of transition boiling. From the data of Witte and Henningson [19], it is found that the temperature for the onset of transition boiling from a sphere immersed in water decreases as the water temperature increases, reaching $\sim 250^\circ\text{C}$ in 60°C water. Since the melting point of tin is 230°C it seems likely that for water temperatures significantly above 60°C , droplets of molten tin would freeze before the onset of transition boiling. Thus the water temperature threshold for explosions

in tin may be understood if it is assumed that the initiation of the first interaction is due to transition boiling.

In general, therefore, it appears from our results that film boiling between molten metals and water at sufficiently high metal and/or water temperatures is stable, but the stability may break down either because of external mechanical perturbation or spontaneously as the surface cools (transition boiling).

It seems likely that the role of the various suggested triggering mechanisms in thermal explosions is merely to disturb the initial stability of liquid/liquid film boiling. It is also probable that the particular perturbing mechanism operating will vary from one incident to another depending on temperature, contacting methods, properties of materials, etc. After the first perturbation, however, the process by which the explosion develops (i.e. by vigorous vapour growth and collapse) appears to be common to a wide range of thermal explosions. Multiple interactions separated by periods in which there are large volumes of vapour have been observed when water is injected into molten salt [20], in water shock tube experiments with aluminium [6], with lead [21] and with salt [21] and in fragmentation studies of a molten metal jet in water [22]. In each case a new interaction occurs when the vapour from the previous interaction collapses. Multiple interactions have also been noted when molten UO_2 is dropped into sodium [23] and when liquid natural gas is poured into water [24].

There is thus some support for the hypothesis that multiple interactions and vapour growth and collapse are generic features of thermal explosions. Since it seems plausible that vapour collapse initiates the subsequent interaction, the mechanism of escalation of a small disturbance into a large explosion is also likely to be generic.

4. JET PENETRATION HYPOTHESIS

It is necessary to consider in what manner the collapse process can initiate a new explosion. Clearly bubble growth is an effective mechanism for converting part of the thermal energy into mechanical energy in the bulk liquid, and there is sufficient mechanical energy associated with a bubble of the size observed to create a large surface area for a new explosion (PV/σ reaches $\sim 1 \text{ m}^2$ for a 1 cm bubble under typical explosion conditions). However, dispersion requires a relative velocity between the two fluids; this is most likely to occur towards the end of collapse when the bubble becomes unstable. The high-speed liquid jet often formed at this stage of

collapse near a wall is known from cavitation studies to be capable of causing significant damage to solid surfaces. The possible role of such jets in producing surface disintegration and rapid energy transfer in metal water explosions has been discussed by Jakeman and Potter [25]. We suggest that if the jet were to penetrate the molten material, rapid fine scale mixing would be inevitable and explosive energy transfer would necessarily follow.*

It is possible to estimate the mixing timescale on the jet hypothesis from the work of Plesset and Chapman [26] who predict that a bubble of the size observed (~ 1 cm radius) collapsing in highly-subcooled water would produce a jet of a few millimetres diameter moving at a velocity of $\sim 10^4$ cm s⁻¹ at the moment of penetration.

If significant mixing is expected after penetration to a few jet diameters [27], then a mixing time of about 10^{-4} s is predicted. This is comparable to the observed explosion pressure rise times.

The degree of dispersal may also be estimated if it is assumed that the inertial breakup of the jet is determined by a critical Weber number [28]. If the penetration velocity is $\sim 10^4$ cm s⁻¹ and $We_{crit} \sim 6$, dispersion of the jet can be expected into droplets of $\sim 3 \times 10^{-5}$ cm radius. Such drops are sufficiently small that energy transfer would be almost instantaneous, even on explosion timescales. The resulting explosion pressure history would therefore be controlled almost entirely by the jet injection rate. It is interesting to note that since the particle size depends on the square of the velocity, the heat transfer time constant, which is proportional to the square of the particle radius, will depend on the fourth power of the injection velocity.

For interactions at low subcoolings, because the jet velocity scales as $(P_\infty - P_v)^{1/2}$, the mixing timescales would increase and the interaction would be expected to be less violent, as is observed. However, the lack of spontaneous explosions above 60°C is unlikely to be due to insufficiently rapid collapse, since $P_v < 0.2$ bar. In fact the impulse-initiated explosions at 80°C ($P_v \sim 0.5$ bar) confirm that 60°C is a threshold for the trigger mechanism only (see above), and that the bubble collapse is still sufficiently energetic to cause escalation. The true explosion threshold must lie between 80°C and 95°C ($P_v \sim 0.8$ bar) at which temperature no explosion could be produced experimentally even with an external mechanical trigger.

* Another possibility is noted: the dispersion is produced relatively slowly during the bubble growth phase and the explosion is initiated by the sudden increase in energy transfer rate which would occur at the moment of vapour collapse.

Most quantitative data on thermal explosions have been obtained from experiments in shock tube geometry; it is therefore particularly interesting to consider whether the same area enhancement mechanism operates in these experiments. It has been demonstrated that there is insufficient kinetic energy in the coolant that enters the interaction region to generate the required dispersal [29]. It can also be shown that if the breakup is determined by inertial forces, and if a critical Weber number ~ 6 applies, then fine dispersal should not occur at the observed bulk column velocities. Consequently, a mechanism is required which can feed kinetic energy from the bulk column into the interaction region and significantly increase the liquid velocities.

Consider therefore the collapse of the last void in the interaction region. When the surface area of the void is much less than the cross-sectional area of the column, the motion of the column will cause rapid acceleration of the void wall, and development of a jet instability as in the bubble case is likely to cause rapid fine-scale mixing if the relative geometric arrangement of void and molten metal is suitable.

More generally, any film boiling situation in which an irregular vapour blanket breaks down is likely to involve at some stage the collapse of void-like regions with consequent localised jet formation. Thus the process of jet formation and penetration is not dependent on the gross geometry of the molten material and coolant. For instance the process could occur effectively when the molten material is in the form of a layer at the base of a vessel (e.g. [30]), or even if the coolant is injected into a sea of hot liquid (e.g. [20]) when it is perhaps more likely that jets of the hot liquid will be formed to penetrate and disperse in the coolant. If the vapour blanket is very thin, as is initially the case with molten metals moving through subcooled water, the scale of the jets and the corresponding depth of penetration will be small, and thus the first interaction will be of limited size, and subsequent interactions would be expected, as is observed.

5. CONCLUSIONS

A number of experiments involving molten tin in water have been performed to investigate dispersion mechanisms in thermal explosions. The thermal explosions recorded usually involve several distinct interactions in the same mass of material, and each interaction is observed to produce a bubble containing some vapour which on collapse initiates the next

interaction. A small interaction may in some circumstances escalate in several cycles of bubble growth and collapse into a large explosion.

The results also indicate that film boiling at the interface between two liquids is unstable to perturbations greater than a certain size. It is suggested that the function of the conventional trigger mechanisms, such as transition boiling (which is shown as the likely mechanism in the case of free-falling drops) or liquid entrapment, is to cause this initial perturbation; the particular mechanism operating may vary from one incident to another, depending on temperatures, contacting methods, properties of materials, etc. The process of cyclic escalation into large scale explosions, however, appears to occur in a wide range of thermal interactions.

It is postulated on the basis of both the present experiments and other work that vapour collapse is the main cause of dispersion in thermal explosions. A possible mechanism for this is that the high-speed liquid jet formed at the later stages of vapour collapse penetrates the molten material and produces a very fine dispersion of the coolant inside it. It has been shown that the jet penetration process can account for both the timescales and energy transfer rates characteristic of thermal explosions, including those in shock tube geometry.

Acknowledgement—This paper is published by permission of the Central Electricity Generating Board.

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FRAGMENTATION DANS LES EXPLOSIONS THERMIQUES

Résumé—On décrit une série d'expériences sur les explosions entre l'étain fondu et l'eau. Les résultats et l'information relative à d'autres travaux indiquent que les explosions thermiques impliquent plusieurs interactions distinctes dans la même masse de matière et que chaque interaction forme une bulle contenant de la vapeur dont la disparition initie l'interaction suivante. On montre ainsi qu'une petite perturbation peut devenir une forte explosion par des cycles successifs de croissance et de disparition.

Il est supposé que la disparition brutale de la vapeur est la cause principale de la dispersion dans plusieurs explosions thermiques. Un mécanisme possible serait la formation d'un jet liquide à grande vitesse (comme observée dans les études de la cavitation) qui pénètre et disperse le matériau en fusion. On montre que l'hypothèse de la pénétration du jet peut considérer à la fois les échelles de temps et les flux de transfert d'énergie, caractéristiques des explosions thermiques incluant celles de la géométrie des tubes de chocs.

ZERFALLSERSCHEINUNGEN BEI THERMISCHEN EXPLOSIONEN

Zusammenfassung—Es wird eine Reihe von Experimenten mit Explosionen zwischen geschmolzenem Zinn und Wasser beschrieben. Zusammen mit den Informationen aus anderen Arbeiten zeigen die Ergebnisse, dass bei thermischen Explosionen gewöhnlich einige bestimmte Wechselwirkungen in derselben Materie beteiligt sind und bei jeder Wechselwirkung Dampfblasen gebildet werden, deren Kollaps die nächste Wechselwirkung auslöst. Damit ist gezeigt, dass sich eine kleine Störung durch sukzessive Bildungs- und Zerfallsprozesse in eine grosse Explosion aufschaukeln kann. Es wird angenommen, dass der Kollaps von Dampfblasen die Hauptursache der Dispersion bei vielen thermischen Explosionen ist. Ein möglicher Mechanismus verläuft so, dass während des Kollapses ein Hochgeschwindigkeits- Flüssigkeitsstrahl gebildet wird (wie in Studien an Kavitationschäden), der in die geschmolzene Materie eindringt und dispergiert. Es wird gezeigt, dass die Strahleindring-Hypothese sowohl die Charakteristik des zeitlichen Ablaufes als auch die der Energieübertragung bei thermischen Explosionen erklären kann, einschliesslich der in Stossrohrgeometrien

ПРОЦЕСС ДИСПЕРГИРОВАНИЯ ПРИ ТЕПЛОВЫХ ВЗРЫВАХ

Аннотация—Описана серия экспериментов по тепловым взрывам между расплавленным свинцом и водой. Результаты настоящих экспериментов, а также данные, полученные в другой работе, указывают на то, что тепловой взрыв состоит из ряда отчетливо различных взаимодействий в одной и той же массе материала и что в результате каждого взаимодействия образуется пузырек, содержащий некоторое количество пара, разрыв которого вызывает последующее взаимодействие. Таким образом показано, что в результате последовательных циклов роста и разрыва пузырьков возмущение может перерасти в значительный взрыв.

Сделано предположение, что основной причиной диспергирования во многих тепловых взрывах является разрыв парового пузырька. Возможно механизм этого явления состоит в том, что во время разрыва пузырька образуется высокоскоростная струя (явление, описываемое в работах по исследованию разрушений, вызываемых кавитацией), которая проникает в расплавленный материал и диспергирует в нем. Показано, что с помощью гипотезы проникновения струи можно учитывать как масштаб времени, так и скорость переноса энергии, которые являются характерными величинами тепловых взрывов включая взрывы в ударной трубе.