AN INVESTIGATION OF THE FRAGMENTATION OF
MOLTEN METALS DROPPED INTO COLD WATER

by

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November 1976

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Reports and Papers Published Under
MIT Fuel-Coolant Interaction Project

(This project was funded until June 30, 1975 by ANL and reports issued as 31-109-38-2831-XX; Starting July 1, 1975 reports were issued as COO-2781-XX).

Progress Reports (Available from National Technical
Information Service, U.S. Department
of Commerce, Springfield, Va. 22151)

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Information Service, U.S. Department
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ABSTRACT

This report represents an investigation to determine the physical mechanism by which small quantities of molten metal fragment extensively when dropped into a pool of cold water. Since this subject has been the focus of considerable research in the past, some of the more prominent theories have been discussed briefly. Experiments were conducted dropping small solid spheres at a high temperature instead of molten metal drops, and indicate a significant difference from the latter. Several hypotheses were proposed based on the hydrodynamics of the molten drop and tested analytically. The theory that the drop fragmentation is caused by the violent release of dissolved gas from within the drop was investigated experimentally and leads to the conclusion that tin fragmentation probably does not occur in this way. It is felt that a calculation of the dynamics of the vapor film that would be expected to surround the hot drop is needed. This calculation was not performed but several suggestions and estimates have been made. It would seem that the possibility of metal fragmentation by rapid vaporization of water entrapped within the metal drop is well worth investigating.

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ACKNOWLEDGEMENTS

I am very grateful for the invaluable help given me by Professor Todreas, Professor Rohsenow and Professor Sonin. I would also like to thank Ken Rhettman for his help in the design of the experimental apparatus.

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NOMENCLATURE

The symbols, explained below, have been used as indicated unless otherwise specified within the text.

a_t	thermal diffusivity
C, c	specific heat
D	diameter
g	acceleration of gravity
h_{fg}	latent heat of vaporization
h_{if}	latent heat of freezing
K, k	thermal conductivity
N_{We}	Weber number
P, p	pressure
ΔP	pressure differential (usually pressure - atmospheric pressure)
∇P	pressure gradient
q''	heat flux
R	drop radius
r	radial coordinate
T	temperature
ΔT	temperature differential
t	time variable
V, v	velocity
X	distance coordinate
	film thickness
	coefficient of viscosity

ρ	density
θ	angle
σ	surface tension
D/Dt	operator denoting substantial derivative

Subscripts

Al	Aluminum
C	cold liquid
C.V.	control volume
h	hot liquid
	liquid
SAT	saturation
Sn	Tin
V,v	vapor
w	drop surface
∞	at infinity or denoting ambient conditions
mpt	melting point

CHAPTER 1

INTRODUCTION

1.0 Introduction

The vapor explosion is a potential problem in a number of industries such as the paper industry, the nuclear industry, the metals industry, to name a few [1], [2], [3], [4], and can lead to damage to both personnel as well as equipment. There is, therefore, a considerable need to develop a proper understanding of the mechanisms which can cause a vapor explosion. This subject has been the focus of considerable research in the past few years, but so far there does not appear to be any consensus on identification of the governing mechanism[s].

The explosive vaporization of a liquid can occur when it comes into contact with a hotter liquid which is at a temperature significantly higher than the boiling point of the first liquid. Based on the exceedingly high vaporization rate, a simple calculation [1] indicates that if 'normal' laboratory heat transfer coefficients are assumed, the initial surface area of contact falls short by several orders of magnitude in accounting for the observed heat transfer rates. It is thus believed by some that this discrepancy may be accounted for by the phenomenon of the fragmentation of the hot liquid which thus makes available

more surface contact area and hence more area for heat transfer to occur. However, it has not yet been established that fragmentation is even a necessary condition for a vapor explosion; and, in fact, fragmentation does not always lead to a vapor explosion. An example of this is the extensive fragmentation that occurs in the experimental test loop when molten uranium oxide comes into contact with liquid sodium. No explosion occurs, but the uranium oxide disintegrates. These test loops seek to simulate the interaction that could occur in a liquid metal fast breeder reactor if molten uranium oxide fuel were to come into contact with coolant liquid sodium. Such accidents are generally known as Fuel Coolant Interactions.

However, fragmentation does provide an explanation of the means of dispersal of a hot molten material throughout a coolant and the consequent area increase for heat transfer in other industries, notably the metals industry.

Thus it is important that the phenomenon of thermal fragmentation be properly understood, and that the mechanisms responsible for it be identified.

1.1 Modes of Fragmentation

Kazimi et al [5] categorised the fragmentation of hot molten materials in coolants according to different modes of contact between the two liquids. These are outlined briefly below.

a) Entrapment Fragmentation

Entrapment fragmentation is believed to be induced by the rapid vaporization of coolant entrapped between the hot molten material and a solid surface such as the bottom of a container which holds the coolant. This mode was demonstrated by the experiments of Long [6] where he found that fragmentation could be prevented by greasing or painting the bottom of the coolant container.

b) Impact Fragmentation

Impact fragmentation is attributed to the effects of the mechanical impact that results when a column of fluid like water held in a tube falls onto hot molten metal at the bottom of the tube, viz. the shock tube experiments of Wright [10].

c) Hydrodynamic Fragmentation

This is believed to be caused by the non-uniformity of the forces resisting the motion of a deformable body in a fluid and is sometimes also called the Weber Number effect since a critical Weber number has to be exceeded for hydrodynamic fragmentation to occur. This critical Weber number is believed to be in the region of 15-22 (Cho et al [7]),

$$N_{WE} = \rho_l V_l^2 D / \sigma_{Sm}$$

for molten tin drops falling in water.

d) Free Contact Fragmentation

Free contact fragmentation, or thermal fragmentation,

is the mode of fragmentation which occurs when small quantities (of the order of a few grams) of hot molten material are dropped into large pools of relatively cold liquid. It cannot be explained by any of the above three other modes of fragmentation. It is this mode which is the subject of this thesis.

1.2 Previously Proposed Mechanisms for the Free Contact Mode of Fragmentation

It is generally accepted that free contact fragmentation is the result of a physical process/processes rather than chemical, since an analysis of the debris has revealed only the presence of the original drop material.

Several physical mechanisms have been put forward to explain free contact fragmentation, but none has so far satisfactorily accounted for all the observed experimental results.

What follows is a brief outline of some of the more prominent hypotheses that have been advanced.

a) Shell Solidification

Herein, it is postulated that fragmentation can occur when the hot molten drop begins to solidify and a freezing front advances inwards from the surface, upon sudden quenching, thereby pressurizing the hot molten core due to shrinkage.

Knapp [8] investigated the problem of a crack in the frozen shell following solidification, and the possibility

of subsequent crack propagation. Since his model was capable of handling only brittle fracture, and cracks were assumed not to propagate in a ductile material, he predicted that brittle fracture of molten tin dropped into water was not possible. However, fracture of uranium oxide contacting liquid sodium could occur in this way since uranium oxide is brittle at lower temperatures.

b) Coolant Entrapment

It has been postulated [9] that when the hot molten drop enters the coolant, a period of unstable film boiling occurs, followed by asymmetrical film collapse, followed by bubble formation and collapse, which leads to a high energy jet of coolant which penetrates the hot drop. This encapsulated coolant then vaporizes explosively within the drop and blows it apart from within. This hypothesis has been refined by Buchanan et al [11] who suggested that the process is cyclic in nature.

c) Spontaneous Nucleation of the Coolant

Fauske [12] suggests that if the molten material is hot enough, the liquid coolant becomes superheated to a degree when spontaneous nucleation occurs and the resulting high localized pressures break the drop apart. 'Spontaneous nucleation' is used to mean either homogeneous nucleation or gas-free heterogeneous nucleation rather than nucleation from vapor-filled cavities which would not exist on a smooth surface such as would be offered by the liquid metal.

Fauske further suggests that perfect liquid to liquid contact must be achieved between the hot and cold liquids, and that the interface temperature must be at or above the spontaneous nucleation temperature threshold of the cooler liquid.

The theory that homogeneous nucleation (nucleation in the bulk of the cooler liquid) is the triggering mechanism for fragmentation fails to explain the fragmentation that has been shown to occur when the interface temperature is below the homogeneous nucleation temperature of the coolant (Appendix 1) Fauske has suggested however that nucleation could occur at the interface between the hot and cold liquids, which he calls spontaneous nucleation. The spontaneous nucleation threshold depends strongly on the contact angle characteristic of the system consisting of the two liquids and the coolant vapor, and would be less than the homogeneous nucleation temperature if the two liquids do not wet each other very well [12] This spontaneous nucleation temperature threshold is therefore lower than the homogeneous nucleation temperature, but still substantially higher than the boiling point of the coolant. However, because of the difficulty of determining the characteristic contact angle, an estimate of the spontaneous nucleation temperature would be difficult to make. .

d) Acoustic Pulse in the Coolant

It has also been suggested [13] that fragmentation

may be associated with the pressure pulse that develops in the coolant because of the sudden heating of the coolant and rise in temperature at the interface with the hot molten material.

Katzimi [5], showed that such a hypothesis was untenable on the grounds that it predicted that the extent of fragmentation would increase proportionally to a temperature difference between the hot and the cold liquid, which does not agree with experimental results.

e) Acoustic Cavitation

It was proposed by Kazimi et al [5] that the pressure fluctuations on the surface of the hot molten material caused by the oscillating film of vapor induce pressure fluctuations within the hot drop, and vaporization could occur from within if sufficiently large negative pressures were produced inside so as to cross the cavitation threshold, thus blowing the hot material apart.

For this calculation [14], the film pressures calculated by Kazimi [5], were used as a driving force on the hot drop surface. It now appears, however, from the experimental work of Bjornard [15] and Bjorkquist [16] that the predicted time scale of interaction was much smaller than that observed.

f) Gas Evolution

Epstein [17] proposed that thermal fragmentation could be a result of violent evolution of gas from within

the hot material caused by the decrease in equilibrium solubility of gases upon quenching the hot material, and the subsequent supersaturation. Our investigation of this hypothesis is discussed in more detail in Chapter [5], and leads to the conclusion that violent gas evolution is not responsible for free contact fragmentation.

So far, the major theories advanced to explain free contact fragmentation have sought to identify a single physical mechanism acting over all ranges of experiment and experience. It is a possibility, however, that not one but several entirely different mechanisms may be responsible, either simultaneously for any given set of experimental conditions, or each separately over different domains of experiment. This could explain, for example, the scatter in experimental results recorded by almost every investigator to date.

1.3 Earlier Experimental Results

Most of the quantitative data taken till this point utilize molten metals such as tin, bismuth, lead, etc. as hot molten materials, with water as the quenching liquid.

High speed movies have been taken of the interactions between molten metals and water by several investigators, e.g. Bjornard [15], Bjorkquist [16] and they indicate that the entire event from the time of immersion into water occupied a time span of the order of milliseconds or tens of ...

milliseconds, depending upon the temperatures of the two liquids. The molten metal drop penetrates the water to some extent, (farther for higher metal temperatures) and then suddenly explodes, scattering debris all around.

As far as the quantitative data available today is concerned, different investigators have used different measures of the intensity of interaction. Thus Cho et al [7] measured the projected area of the original drop and compared it with the projected area of the fragments, Armstrong et al [18] measured the number of fragments, Buchanan et al [11] measured the percentage disintegration which they define as the ratio of the weight of the fragmented portions to the weight of the unfragmented residue. Bjornard [15] and Bjorkquist [16] measured the pressure pulses in the water, during the event, by means of a pressure transducer placed in the water a little distance away from the interaction and used the peak pressure magnitudes as a measure of the intensity of interaction.

1.4 Factors Affecting Extent of Interaction

So far the following factors have been identified as being very important in determining the intensity of the fragmentation interaction.

- a) Identity of the two liquids
- b) Temperature of the hot liquid
- c) Temperature of the cold liquid
- d) Quantities of the two liquids involved

a) Effect of Identity of the Two Liquids

A lot of molten metals will fragment in the free contact mode when dropped into water within a given temperature range. Figure (1) taken from Ref. [34] shows this behavior. Some metals will fragment more extensively than others in water under nominally the same conditions; the identity of the coolant also decides the extent of fragmentation, if any, (McCracken [19]). However, the particular thermal, fluid or any other properties have not yet been individually isolated as being significant.

b) Effect of Temperature on the Hot Liquid

The data of Chio et al [7] indicates that for metals such as tin, the extent of fragmentation depends very strongly on the initial temperature of the molten metal prior to dropping into cold water. Fragmentation usually begins with the metal initial temperature is somewhat higher than the melting point of the metal, increases with increasing metal initial temperature over a range, and then begins to decrease as the metal temperature is increased further, all this while the water temperature is kept constant as shown in Fig. (2) from Chio et al [7]], when the reversal is seen to occur at about 600° C for tin dropped into water.

This behavior is borne out by the experiments conducted by Bjorkquist [16], see Fig. (3). The values reported by Bjorkquist are values calculated to exist at the drop

surface at the instant of fragmentation from values measured experimentally a distance away. The original values of the pressure recorded were calculated and are indicated in Fig. (4).

The dwell time, defined to be the time between entry of the molten metal drop and fragmentation, is observed in Fig. (5) from Bjorkquist [16] to increase monotonically with increasing initial metal temperature. Thus at higher metal temperatures the molten metal drop penetrates further into the coolant pool before fragmentation. This is believed to be attributable to the fact that a film initially exists on the drop surface which collapses when the drop temperature falls to a value where it can no longer support the film, and that film collapse must occur before fragmentation is possible. Thus a drop at a higher initial temperature of dropping would take longer to cool down below the minimum film boiling point, and hence a longer time for fragmentation to begin would be the result.

c) Temperature of the Cold Liquid

Quantitative experiments appear, at this time, to be restricted to molten metals dropped into water. The experiments of Cho et al (Fig. 2) indicate that the extent of fragmentation decreases as the water temperature is increased or equivalently as the degree of subcooling decreases. The rather limited (in this respect) experimental results of Bjornard [15] also suggest this behavior.

More recently, Dullforce et al [20] have identified a region they call the temperature interaction zone within which interaction, i.e. free contact fragmentation occurs, Fig. (6). Outside this zone, no fragmentation is possible. Thus there appears to be a cutoff temperature as far as the coolant is concerned, above which there is no fragmentation. That such a cutoff temperature does indeed exist has been indicated by others as well, e.g. Cho et al [7] although the work of Dullforce et al has greatly extended the knowledge of the variation of this temperature as a function of bulk coolant temperature.

d) Quantity of the Two Liquids Interacting

We consider only the case of hot molten metal drops, of weight of the order of a few grams, dropped into a large pool of water. There does not appear to be any quantitative data available at this time, but it has been reported that increasing the molten metal drop size tends to increase the violence of interaction.

Note: Bjorkquist [16] used two different drop masses 0.8 g, 6.6 g in his experiments and reported that the peak pressures he calculated were significantly greater for the larger drops. These values were however those calculated to exist at the drop surface during the interaction from the pressure signals he picked up and recorded using a pressure transducer placed some distance away in the water. However, later experiments, discussed in Chapter 2, now

indicate that there is some doubt as to the formula employed in the calculation. When one employs the actual pressures measured at the transducer face, the magnitudes of the peak pressures are not much different from the two drop sizes used. Experiments using tin drop masses of 2 g. are reported in Chapter 5.

Almost every investigator has reported a fair amount of scatter in the experimental results from laboratory dropping experiments. It is possible that some factor, not considered in the above discussion has been overlooked and not maintained constant throughout a given set of experiments, thereby causing the large scatter in data. However, by considering the average value of the results, the general trends in behavior discussed previously seem fairly clear.

CHAPTER 2

EXPERIMENTAL EXTENTION OF BJORKQUIST'S WORK

2.0 Introduction

This chapter deals chiefly with the experimental work done employing the same apparatus used by Bjornard [15] modified by Bjorkquist [16]. Proper functioning of the apparatus was investigated as well as the formula utilized by both Bjornard and Bjorkquist to compute the pressure at the surface of the drop from the experiments they conducted. In addition similar experiments were run with solid steel balls instead of molten metal, though no movies were taken. The apparatus, which is described more fully by Bjornard, is reviewed below and illustrated on Figures (7) and (8).

2.1 Details of the Experimental Apparatus and the Experiment

The drop material is placed inside a small crucible which is then lowered into a vertical tubular furnace, placed several inches above the coolant pool surface. The metal temperature is measured with a platinum rhodium thermocouple connected to a pyrometer readout. The sample is heated in the center of the furnace to a temperature slightly above that at which it is to be dropped while argon gas is passed through the furnace. It is then lowered to the bottom of the furnace just above the coolant pool, and dropped by pressing

a button that activates the solenoid release. This causes the driver spring to turn the rotating cam and with it a rotating shutter plate which is below the drop inside the crucible. A hole in the shutter plate lines up with a hole in the base of the crucible and the drop falls through into the water.

The pressure pulses inside the water are picked up by a pressure transducer, and twenty milliseconds of the interaction are recorded by a taperecorder on a small loop of magnetic tape which may be played back at any desired time onto an oscilloscope.

The electronics circuit is shown in Fig. (8). As the drop falls from the crucible it cuts a light beam just before entering the water. This generates a pulse from an optical trigger circuit which is used to trigger an oscilloscope. This trigger may then be transferred to the signal recorder after an appropriate time interval by means of the delay-time trigger transfer feature on the oscilloscope. This delay time is useful since the recording capacity of the recorder is only twenty milliseconds, and it ensures a timely 'window' of the pressure pulse during the actual interaction.

The drops are collected below in a cloth cap attached to a stick to facilitate easy insertion and removal from the tank.

2.2 Experimental Precautions and Verification of Accuracy

In order to check the possibility that the natural frequency of the transducer interfered with the observed frequency, the response characteristics of the transducer system were measured. A pure step rise in pressure produced by means of a pulse applied by a shock wave in gas was used, and the transducer system response was noted on a photograph.

The rise time of the transducer is less than one millionth of a second and the resonant frequency of the transducer is estimated to be 400 kilohertz, which damps out in about 200 microseconds. The observed frequencies in the experiments are 20 Khz at most, and since these are very much less than the natural frequency of the transducer, the interference of the transducer characteristics is expected to be insignificant.

2.3 Calculation of Pressure at the Drop Surface

Bjornard measured the pressure behavior in the water during fragmentation. He recorded the peak pressures and called it the "Big Bang," since this region was characterized by a relatively low frequency high pressure pulse and thought to coincide with fragmentation. From these peak values he employed a formula to calculate the pressure at the drop surface $p(a)$.

$$p(a) = \frac{(TD - \bar{V} \Delta t_d)}{a} p(r) \quad (1)$$

where

TD is the transducer depth

$\bar{V}$ is the average droplet velocity

Δt_d is the dwell time

a is the sphere radius

p(r) is the recorded pressure at the transducer

In order to verify that the above formula did indeed hold, experiments were performed dropping small masses of molten tin into water and measuring the pressure field at different distances from the drop surface. Two transducers were employed simultaneously at differing distances and each one was connected to a different signal tape recorder to eliminate any scatter from the variability of the generated pressure in sequential experiments. The results were inconclusive and though most of the data points fell evenly around the line represented by equation (1) the scatter was too high to permit continued use of the formula. For this reason the results of Bjornard and Bjorkquist (who used the same formula) are being presented here through the recorded pressures and not the calculated pressures. The experimental data is shown in Appendix (4).

2.4 Experiments and Results

Experiments, as indicated in 2.1, were conducted using large solid spheres instead of the molten metal drops employed by Bjornard and Bjorkquist in order to see what was the

FIG. 9: SKETCH OF PRESSURE TRACES

difference in pressure behavior between a liquid-liquid system where fragmentation was observed to occur and a solid-liquid system where no fragmentation occurred. These solid spheres were made of steel* and were of diameter 8 mm.

The pressure behavior is sketched in Fig. (9). The 'Big Bang' reported by Bjornard [15] is noticeably absent, and though there is a peaking of pressure and a 'tail', it is much more gradual and far less pronounced than if molten tin had been used. The pressure behavior is thought to represent an oscillating film of vapor at the drop surface, a condition of unstable film boiling or transitional boiling.

The peak pressures are indicated in Fig. (10) as a function of the initial drop temperature.

Frequency analysis of the pressure recordings was done using a digital spectrum analyser. Here another difference manifests itself in that the frequency spectrum is fairly uniform, and the dominant frequencies reported by Bjornard for the molten metal - water system are absent.

It should be noted that the drops would turn a light grey on the surface--probably because of the formation of oxide--even though argon gas was passed through the furnace during the heating of the drop.

The important conclusion that can be drawn from these experiments is that the 'Big Bang' is characteristic of

*Specifically CR52, a chrome steel used as a bearing material.

fragmentation in a liquid-liquid system, and that the 'Big Bang' peak pressure is therefore a good measure of the intensity of the interaction.

2.5 Multiple Small Drops

The experiments described above were then repeated using a number of smaller steel drops simultaneously, each of diameter 4 mm. Eight balls were dropped at a time and the pressure pulses recorded. The purpose was to compare the pressure fluctuations, at the point of peaking, with one drop.

The pressure traces were more random here, as might be expected, and the scatter of data was larger as Fig. (9) shows. A number of pressure peaks were noticed, as opposed to the single peak of single drop experiments, due probably to each drop reaching a peak at a slightly different time.

Figure (11) shows the peak pressure as a function of the initial temperature of the drops.

2.6 Summary and Concluding Remarks

It is evident that the pressure behavior traces recorded for the solid drops is significantly different from that of the molten drops, notably the absence of the 'Big Bang.' Although the pressure peak magnitudes are different for the solid drop, this could be accounted for by the different thermal properties of the different materials used.

The frequency spectrum of the solid drops is much

more uniform than those of the molten drops which exhibit two dominant frequencies.

A lot of scatter is noted in the data as with the molten tin drop data. This could mean the presence of a factor not accounted for by the thermal processes, i.e. fluid mechanics and heat transfer of the event.

CHAPTER 3

HYDRODYNAMIC EFFECTS CONSIDERING THERMAL CONDITIONS

3.0 Introduction

This chapter concerns itself chiefly with theories that try to correlate the extent of fragmentation experimentally observed with a characteristic parameter of the corresponding experiment or a theoretical model such as the peak pressure observed on the recorded tapes of Bjorkquist [16], a Weber number, a time integrated pressure response, etc.

Although the results were admittedly not very encouraging, they do, however, serve to provide a test of the different possibilities set down here.

3.1 The Initial Weber Number Model

When a molten drop of tin or bismuth falls into a coolant (e.g. water at room temperature) it undergoes an interaction with the coolant resulting in a break-up of the drop; at the same time, the pressure at the surface of the drop undergoes oscillations of high frequency.

In this model, where the drop falls through the water at a few feet per second, the vapor film surrounding the drop is thought to be flowing past the drop, upwards. Separation occurs on the upper surface of the drop which is therefore a low pressure region. It is suggested that at this point the pressure is relieved close to atmospheric pressure. There

is thus a non-uniformity of pressure forces at the surface of the drop which would tend to break up the drop. Resistance to this drop breakup is offered by the surface tension of the molten metal drop.

Thus a Weber number is defined such as,

$$N_{We} = 2 (\Delta P) D / \sigma \quad (2)$$

with $2\Delta P$ replacing the more usual ρV^2

where

ΔP is the peak pressure minus atmospheric pressure

D is the drop diameter

σ is the surface tension of the liquid drop with air
is not expected to change the substantially.

The P in eq. (2) was estimated from the values of the peak pressure predicted by Kazimi [5]. It was sought to correlate the Weber number with the experimentally determined extent of fragmentation recorded by Cho et al [7] as shown in Fig. (12) and Fig. (13). The experimental data of Bjornard [15] was not used because the N_{We} would just be a constant times the ΔP calculated by Bjornard since σ and D are nearly constant, and an attempted correlation would just reduce to a plot of ΔP (used as a measure of the intensity of interaction) determined by Bjornard, against a N_{We} which would be just a constant times the ΔP . The data of Cho et al on the other hand characterizes the interaction intensity

as a function of measured final surface area rather than pressure and thereby avoids this problem.

The N_{We} is seen to increase almost linearly with initial drop temperatures in contrast to the experimental fragmentation curve which rises initially, reaches a peak, and then falls again. The behavior of fragmentation as a function of pool temperature was similarly investigated as Fig. (13) shows. The graph of N_{We} against coolant temperature also does not agree with the experimental fragmentation curve.

In Fig. (12) and Fig. (13), peaks 1 and 2 indicates that N_{We} is based on the first and second peak pressures respectively in the first two cycles of the oscillation of the film pressure predicted by Kazimi.

It has been reported, Cho et al [7], that the critical Weber number for hydrodynamic breakup is in the region of 13 to 22 for tin drops falling into water. This Weber number is defined in the customary way:

$$N_{Ww} = \rho V^2 D / \sigma \quad (3)$$

ρ is the density of the coolant (water)

V is the drop velocity

D, σ as defined in equation (2)

In each case, the Weber number calculated by equation (2) computed was much higher--in fact, a simple calculation indicates that a pressure difference of only about 0.1 psi across the drop is sufficient to cause break-up.

3.2 The Final Weber Number Model

As the molten metal drop undergoes a thermal interaction with the colder water, the Weber number as defined by equation (2) is considerably higher than critical at first, and hence here the drop is assumed to break up into smaller droplets. Each droplet thus formed then is assumed to undergo a similar interaction and breaks up even further because of the high non-uniform vapor pressures it is subjected to. This process repeats itself cyclically until the Weber number decreases to a critical value, perhaps the value given by Cho et al [7] for tin and water, i.e. 13-22, at which point further breakup terminates. Thus the final metal fragment size is determined by the final critical Weber number. From the data of Cho et al the final diameter of the fragment was calculated for the different experimental runs reported, and used to run Kazimi's [5] computer program to predict the pressure history of the drop as it enters the last interaction cycle and the drop size reaches its final smallest value, i.e. Kazimi, in his calculations used drop diameters previous to quenching; here, the final drop sizes of the fragments were used. Since the temperature of the drop and other thermal conditions are not known at the final breakup stage, they were approximated by conditions at the outset of the whole experiment, i.e. by assuming that no cooling had occurred till then.

The peak pressures predicted with this calculation were then used to define a Weber number similar to the one given by

equation (2),

$$N_{We} = 2(\Delta P)D/\sigma$$

It was sought to show that this N_{We} would be a constant equal to a critical value. The results of this calculation are shown in tabular form in Fig. (14). It is readily seen that N_{We} does not remain constant, thus indicating that the mechanism of breakup is not as proposed above.

It is to be noted that owing to the general spread in the fragmentation data, and also to the fact that a unique final fragment diameter is not realistic but has mainly statistical significance in being an average, the results indicated above are very approximate.

3.3 The Time Integrated Pressure Model

This model suggests that the criterion or key parameter in drop fragmentation is not just pressure per se, but rather a pressure 'impulse' or pressure integrated over a length of time, i.e. when the molten metal drop is subjected to the pressure of the coolant vapor, this pressure acts over a finite interval of time which is also important to the process. It was therefore sought to correlate a characteristic time-integrated pressure value of the process with the fragmentation data.

Figs. (15) and (16) are based on Kazimi's prediction of the vapor pressure history. The integrated pressures indicated are calculated over a time period of 10^{-4} seconds starting from

time 10^{-7} seconds which is the length of time Kazimi ran his calculations. Fig. (15) for varying initial drop temperatures shows a fairly good correspondence, but Fig. (16) for varying coolant temperature does not.

The film pressure predicted by Kazimi oscillates about the atmospheric pressure. In Fig. (15) and Fig. (16) the +ve curve is a calculation of the time-integrated value of the difference between the film pressure and atmospheric pressure always considered positive. The - ve curve is a calculation of the time-integrated value of the film pressure minus the atmospheric pressure, considered positive only if the film pressure exceeds atmospheric and considered negative otherwise.

Although the calculations were based on a time span of 10^{-4} seconds, the shape of the curves in Figs. (15) and (16) are not expected to change much because the pulsations predicted by Kazimi die down at this point, and the shape of the curves would be independent of the number of cycles used in the calculation.

The calculations were then repeated using the pressure histories recorded on tape by Bjornard (15). The peak pressure (Bjornard called it the 'Big Bang' pressure) amplitude with the 'Big Bang' frequency recorded yielded a pressure 'impulse' over one half-cycle. Similarly, there is a time integrated pressure associated with the high frequency portion of the pressure signal recorded by Bjornard. Figs. (17) and (18) show the results of the integrated pressure. Again, the

agreement with the fragmentation data is not good.

3.4 Summary and Concluding Remarks

In this chapter surface tension mechanisms, i.e. mechanisms for fragmentation in which the breakup is caused by pressure forces overcoming the resistance offered by surface tension, were modelled but with negative results. In addition, a comparison of a time integrated pressure with the fragmentation data was attempted but proved to be unproductive.

CHAPTER 4
ON SOME ASPECTS OF THE DYNAMICS
OF THE FILM SURROUNDING THE DROPS

4.0 Introduction

As the hot molten drop of metal comes into contact with the coolant a number of events take place. On impact with the coolant surface the drop flattens somewhat and there is a distinct wake behind it. If the initial drop temperature is high enough, film boiling is expected to occur over the leading portion of the drop and high speed photographs taken by Bjorkquist [16] clearly show vapor films around the drop which eventually detach and rise through the water when the drop is initially hot enough. At lower drop temperatures nucleate boiling may be expected and the fragmentation occurs almost instantaneously.

It has long been suspected by many investigators that this film of vapor around the drop is somehow responsible either directly or indirectly for free contact fragmentation, e.g. Witte et al [21], Kazimi [5].

The film around the drop is thought to pulsate as a result of the rather complicated film dynamics and heat transfer of the interaction. This vapor film pulsation has been reported experimentally by some investigators, Walford [22], Stevens [23], Bjornard [15] and Bjorkquist [16].

It has been proposed that the fragmentation can result from the pulsation of the vapor film in any of the following ways:

a) Associated with the film thickness pulsation there is also a pulsation of the pressure that the film exerts on the surface of the drop. This pressure is not exerted symmetrically around the drop so that when high enough pressures are reached in the vapor film, owing to the non-uniformity of these pressures, the drop breaks up. Thus the appropriate criterion for fragmentation under this hypothesis would be the magnitude of the pressure generated in the film and the difference in pressure across the drop.

b) Another postulate suggests that the pulsations of the vapor film eventually die out and the film collapses locally onto the molten metal surface. By some means, not definitely identified, if a bubble were to form on the drop surface, expand and then collapse, a high velocity jet of coolant might then result directed towards the drop so as to cause extensive fragmentation by entering the drop and then vaporizing explosively from within. This process could repeat itself cyclically and result in extensive breakup in a very short time span, e.g. Buchanan et al [11].

c) Yet another hypothesis advanced by Kazimi et al [5], suggests that as the vapor film pressure oscillates, it induces similar pressure fluctuations inside the molten

drop surface. Thus a 'negative' pressure fluctuation could also occur in the drop interior which would be magnified, thus facilitating internal cavitation leading to fragmentation from Kazimi et al [5]. (Cavitation denotes the process of growth of vapor nuclei within a liquid when the liquid pressure is reduced.)

Many investigators have expressed the opinion that vapor film collapse and the consequent liquid-liquid contact are necessary conditions that must be satisfied before any fragmentation can take place. However, without going into a complicated calculation of the film dynamics of the drop it becomes difficult to predict, with much confidence, any kind of minimum film boiling point.

Henry's correlation [26] for the minimum film boiling point is based on two effects, namely a) intimate contact between the two surfaces b) microlayer evaporation, giving rise to surface cooling transients. While the correlation is successful for liquid-solid systems, there is some doubt as to its applicability for liquid-liquid systems, as demonstrated by Henry et al [33] who suggest that if the two liquids do not wet well, the microlayer evaporation term might not apply.

From the foregoing it appears that an accurate prediction of the heat transfer from the drop and the dynamics of the film around the drop may be desirable to accomplish at

some time in the future. Although the calculation has not been accomplished to date at M.I.T., a few points of interest that may be useful have been studied and are discussed in the following sections. A model is also outlined briefly in Appendix 5.

4.1 Dynamic Film Behavior with Time

A detailed analysis of the dynamic film behavior with time is not an easy task and usually simplifying assumptions have to be made which may not always be realistic. A major problem in such an analysis is the explanation of the existence of a film or its creation, i.e. the mode of generation of vapor at a liquid-liquid interface, apparently devoid of preferred nucleation sites in the nature of vapor-filled cavities, without spontaneous nucleation. This is usually circumvented by postulating an initial vapor or gas film thickness to exist, possibly air carried in by the drop as it enters the water. A second problem then arises as to a good or even realistic estimate of this initial film thickness.

Kazimi [5], postulated that the initial film thickness was of the order of 10^{-5} cm or 10^{-6} cm (specifically for a molten tin sphere of radius 0.3 cm. dropped into water). He also made the assumption that the interaction was spherically symmetrical in nature and neglected any flow in the vapor past the sphere which may seem rather difficult to justify in that high speed photographs subsequently

obtained by Bjornard and Bjorkquist do show streaming flow past the sphere. A problem seems to be that the calculated pressures in the film change a good deal depending upon the initial film thickness chosen to start the calculation.

Hsaio, et al [21], developed an analytic model to predict the sequence of events associated with the hot molten metal sphere dropped into a colder liquid. A vapor film is assumed to form instantaneously on contact, the solution being invalid at time $t=0$. The vapor surrounds and flows past the sphere so that a significant variation is found in film thickness with the angular position. No pressure calculations were made. The choice of a steady velocity field in the coolant liquid and the absence of a radial momentum equation, however, seems to have eliminated the vapor film oscillations predicted by Kazimi, and the calculation does not proceed to film collapse.

4.2 An Estimate of the Film Thickness

The problem of estimating the film thickness has been pointed out in the previous section. An attempt was made to estimate it in the following way.

As Fig. (19) shows, the drop is falling down through the water or equivalently the water is imagined to be flowing upwards past the drop. Separating the two liquids is a film of vapor surrounding the drop and also flowing past the drop. An order of magnitude estimate is attempted

and we may consider a kind of 'steady-state' estimate when the amount of vapor being generated by vaporization of the coolant liquid is just balanced by the amount of vapor that leaves the film by flowing past the drop. Since this is not a calculation of any detail, any dependencies on angular position, etc. are ignored.

If a linear temperature profile is assumed in the vapor film, the heat flux to the coolant interface is given by,

$$q^* = K_v (\Delta T) / \delta \quad (4)$$

If all of this heat flux goes into vaporising the coolant liquid, the rate of production of vapor over the leading half of the drop may be roughly estimated to be:

$$\omega_v = K_v \left(\frac{\Delta T}{\delta} \right) \cdot \left(\frac{2\pi R^2}{h_{fg}} \right) \quad (5)$$

This vapor production rate is equal, in our steady state estimate, to the rate of flow of vapor out of the film,

$$\omega_{flow} = 2\pi R \delta \rho_v u_v \quad (6)$$

and

$$\frac{K_v (\Delta T)}{\delta} \cdot \frac{2\pi R^2}{h_{fg}} = 2\pi R \delta \rho_v u_v \quad (7)$$

Our unknowns are δ and u_v . We next establish a second equation in these unknowns by examining the pressure balances in the film along the direction of the flow.

As the vapor and coolant liquid flow up past the drop, separation is expected to occur over the rear portion of the drop and the pressure over the rear half is therefore close to the ambient pressure. By assuming that the flow of the liquid coolant is potential or inertia-governed the pressure difference between the leading point of the drop and the rear portion is given to an approximation by,

$$\Delta P_L \approx \rho_L u_L^2 / 2 + \rho_L gR \quad (8)$$

Since the film is expected to be very thin, the pressure of the liquid may be considered to be impressed upon it. The pressure difference balances both the viscous shear and the inertia of the vapor. By imagining the flow to be like the flow between parallel plates the first term may be estimated to be,

$$\Delta P_f \approx 12 \frac{\eta_U u_U}{\delta^2} \cdot \pi R \quad (9)$$

The second term is approximated to be

$$\Delta P_m \approx \rho_U u_U^2 / 2 \quad (10)$$

Then equating the pressure drops in the liquid coolant and the vapor the following equation is obtained:

$$12 \frac{\eta_U u_U}{\delta^2} \cdot \pi R + \rho_U u_U^2 / 2 = \rho_L u_L^2 / 2 + \rho_L gR \quad (11)$$

Equations (7) and (11) may now be solved by a process of trial and error.

For a molten tin drop falling in water, the values used below are representative of laboratory conditions:

$$R = 0.01 \text{ ft.}$$

$$v_{\ell} = 3 \text{ ft. per sec}$$

$$\Delta T = 400^{\circ}\text{F}$$

This gives $v_u = 9.3 \text{ ft/sec}$ and $\delta = 2.2 \times 10^{-4} \text{ ft.}$

This 'steady' estimate of δ may be regarded in the nature of an upper limit of δ , since if δ were to become larger, the heat flux and consequently the vapor generation rate may be expected to decrease in accordance with eq. (5) and the vapor mass leaving the film to increase in accordance with eq. (6) and eq. (11).

4.3 An Estimate of the 'Big Bang' Frequency

As reported earlier, the instant of fragmentation is accompanied by a high pressure, comparatively low frequency trace in the pressure recordings which were made by the transducer. High speed photographs also clearly show several pulsations at the moment of fragmentation much like a mini-explosion. It was suspected that the pulsations were caused by the rapid oscillations of the high pressure vapor suddenly generated and not due to separate interactions occurring at the same location. The rough order of magnitude calculation indicated below seems to support this view.

At the instant of fragmentation, when the high pressure vapor is generated, the system is modelled as a mass spring

system. The inertia is supplied by the surrounding coolant liquid, and the spring stiffness is supplied by the compressibility of the vapor.

As the liquid pulsates around the vapor, we may write, for a spherically symmetric liquid motion radially,

$$\rho \frac{\partial v}{\partial t} = -\partial p / \partial r \quad (12)$$

Also,

$$4 \pi r^2 v = 4 \pi R^2 v_0 \quad (13)$$

where v_0 is the velocity at the interface from the conservation of mass in the incompressible liquid. Thus,

$$\frac{\partial v}{\partial t} = \frac{R^2}{r^2} \frac{\partial v_0}{\partial t} \quad (14)$$

$$v_0 = dR/dt \quad (15)$$

$$\rho \frac{R^2}{r^2} \frac{d^2 R}{dt^2} = -\partial p / \partial r$$

Integrating this between $r = R$ and $r \rightarrow \infty$

$$P_\infty - P_0 = -\rho R^2 \frac{d^2 R}{dt^2} \int_R^\infty \frac{dr}{r^2} = -\rho R \frac{d^2 R}{dt^2} \quad (16)$$

To obtain the equivalent spring stiffness of the vapor, it is assumed that the vapor undergoes an adiabatic process reversibly. This assumption is, of course, not correct but it will serve for a very rough calculation such as the one

being attempted here.

With

P_o = pressure in the film

V_o = volume of vapor in the film

γ = adiabatic gas constant

We may thus write:

$$P_o V_o^\gamma = \text{constant}$$

$$P_o V_o^\gamma = P_\infty V_i^\gamma \quad \text{when the film expands till } P_o = P_\infty \text{ and} \\ \text{radius } R_i.$$

Since $V \propto R^3$

$$P_\infty = P_o (R_i/R)^{3\gamma} = -\rho R \frac{d^2 R}{dt^2} \quad (17)$$

$$1 - (R_i/R)^{3\gamma} = -R_i^2 \frac{\rho R/R_i}{P_\infty} \frac{d^2 R/R_i}{dt^2} \quad (18)$$

Use of the binominal theorem gives,

$$\epsilon (3\gamma) = - \frac{R_i^2 \rho}{P_\infty} R/R_i \frac{d^2 R/R_i}{dt^2}$$

$$\text{where } R_i/R = 1 + \epsilon \quad \epsilon \ll 1$$

If we choose a scaling time such that

$$\frac{d^2 R/R_i}{dt^2} = \frac{R/R_i}{\tau^2}$$

$$\tau = (\rho R_i^2 / P)^{1/2}$$

$$\tau \approx R_i \sqrt{\rho/P_\infty} \quad (19)$$

Substituting

$$R_i \approx 0.5 \text{ cm}$$

$$P_\infty \approx 14.7 \text{ psia}$$

$$\rho \approx 62.5 \text{ lb/ft}^2$$

then

$$\tau \approx 5 \times 10^{-4} \text{ sec}$$

Or equivalently a frequency of $f \approx 1/2 \approx 2000 \text{ hz.}$, which is of the same order of magnitude as the 'Big Bang' frequency reported by Bjorkquist [16].

4.4 Velocity of Coolant Liquid Jet

It has been suggested earlier that fragmentation may be caused by the local collapse of the vapor film surrounding the drop, the formation of a bubble, its collapse, the formation of a liquid coolant jet directed towards the drop, the consequent mixing of the jet and the molten metal, and the explosive vaporization of the liquid coolant from within the drop.

A rough estimate was made of the speed of such a coolant jet should it occur.

As noted in 4.3, the radial velocities in the liquid coolant are expected to die out within a radial distance of

the drop equal to one drop radius, because the vapor film thickness is expected to be much smaller than the drop radius. Since the jet is driven by the pressure difference between the ambient liquid pressure and the low pressure in the collapsing film, this pressure difference is approximated by the magnitude of the ambient pressure which is close to atmospheric pressure P_{∞} .

$$\Delta P \approx P_{\infty}$$

For the liquid motion we may write

$$\rho \frac{Dv}{Dt} \approx - \partial p / \partial x$$

$$P_{\infty} / R \approx \rho v^2 / R$$

where v/τ is the acceleration of the liquid and $\tau = R/v$

Thus giving

$$v = \sqrt{P_{\infty} / \rho}$$

For $P_{\infty} = 14.7$ psi

$$\rho = 62.4 \text{ lb/ft}^3$$

Then $v \approx 33$ ft/sec is an estimate of the velocity of the liquid jet, thereby indicating that the jet theory of coolant penetration into the metal may be well founded.

4.5 Time for Deformation of Drop

A point of interest in the analysis of the dynamic film behavior is whether the molten metal sphere remains a

sphere or is appreciably deformed when it enters the water.

The following order of magnitude calculation was performed to investigate this possibility.

We estimate the time needed for the molten metal to deform through a distance equal to the sphere radius, at any point,

For this deformation we may write, approximately,

$$\rho \, Dv/Dt \approx \nabla p$$

where

ρ = molten-metal drop density

v = velocity of deforming metal

t = time variable

p = pressure

D/Dt = operator indicating the substantial derivative

∇p = pressure gradient in metal which causes the acceleration Dv/Dt

Let τ = time required for the deformation

Then a representative velocity of the metal, v , would be given by the distance of deformation R divided by the time taken for the deformation τ .

$$\text{i.e. } v \approx R/\tau$$

The pressure gradient ∇p may be approximated by the pressure peak magnitude of the film divided by a distance equal to the distance of deformation.

$$\nabla p \approx (\Delta P)/R$$

Thus we may write,

$$\tau = \sqrt{\rho/(\Delta P)} \quad R$$

Thus for a molten-tin drop falling in water, times needed to appreciably deform the drop were computed, using Bjornard's results to estimate P at the drop surface, for want of a better estimate. The following values were used:

$$\rho = 424 \text{ lbm/ft}^3$$

$$R = 0.01 \text{ ft}$$

The results are indicated on Fig. (20). Based on this calculation it would appear that deformation of the drop during its fall is significant, within the time scale of the experiment. For example, at a temperature of 500° C, when the dwell time is about 8 m sec., the time to deform the drop appreciably is almost 0.5 msec.

4.6 Summary and Conclusions

A rough calculation indicates that the amount of deformation of the drop as it falls through the water is probably very significant.

The rough estimate of a steady film thickness, 2.5×10^{-4} feet should serve as an upper bound for future calculations. In addition, the vapor velocity in the film was estimated to be in the region of 10 feet per second. Both estimates were made for conditions matching those in the

laboratory experiments.

An estimate of the speed of a liquid jet of coolant striking the drop was made, and such a high velocity jet could well be an important factor in the drop fragmentation.

An estimate of the 'Big Bang' frequency suggests that the pulsations are probably due to the compressibility of the vapor around the drop and the inertia of the liquid coolant and not due to separate interactions occurring at the same locations.

CHAPTER 5

GAS EVOLUTION AS A MECHANISM FOR FRAGMENTATION

5.0 Introduction

Fast [24] proposed that gas evolution was responsible for the porosity observed in a weld. Epstein [17] using this as a basis proposed that free-contact fragmentation of a molten metal dropped into a coolant liquid is caused by the violent release of dissolved gas from within the molten metal. His theory is outlined in detail below.

The equilibrium solubility of gases in molten metals increases with both temperature and pressure, in general. Thus when the molten metal is cooled very rapidly, it becomes supersaturated and expels dissolved gas. Epstein suggested that this gas release occurs violently and in doing so causes the drop to break up. He pointed out that although homogeneous bubble nucleation theory implies that latent pressures of 10^6 atmospheres or larger are needed to nucleate a gas bubble, experimental evidence suggests a latent pressure of about 100 atmospheres is sufficient, e.g. silver-oxygen system. He also showed by a simple calculation that the gas release mechanism seems to account for the time scale of fragmentation.

5.1 Theory

Diatomic gases that do not react with the metals such

as hydrogen, nitrogen, or oxygen dissolve, ideally, in accordance with Sievert's law:

$$C_o = A P_o^{1/2} \exp \left(- \frac{\Delta h_o}{RT} \right) \quad (20)$$

where

C_o = equilibrium mass fraction of dissolved gas

A = constant

P_o = partial pressure of gas above solution

R = universal gas constant

T = temperature of system

Δh_o = positive enthalpy change across the dissolution reaction e.g., $1/2 O_2 (g) \rightarrow O (diss.)$

Figure 21 illustrates this behavior.

Thus if a molten metal drop were heated for a long time in atmospheric air, say at 500°C, the partial pressure of oxygen above the metal would be 0.2 atmospheres. If the drop were suddenly quenched in water to say 420°C so rapidly that no oxygen could escape by diffusion, then on Fig. 21 one would proceed to point B1 from point A1.

Alternatively, one could consider the driving force to be the amount of supersaturation at state B2 (Fig. 21) by considering actual pressures rather than latent pressures.

The pressures of states A1 and B2 could then be considered to be externally imposed and the system would then follow a path such as A1 to B2 depicted in Fig. 21. A

measure of the driving force for fragmentation would then be the amount of supersaturation or the difference in concentrations between the states A1 and B2. There is a problem, however, in determining the pressure of the gas when the drop is immersed in water and the second state B2 may not be as shown in Fig. [21]. For this reason we adopt the latent pressure as the measure of the driving force from here on.

If the experiment was started with the molten metal sample not quite saturated, then the driving force for fragmentation would be less. A family of curves may be generated as shown in Fig.[22] to allow for the situations where the molten metal sample has an amount of gas dissolved equal to a fraction of the equilibrium concentration. For the ideal system, the driving force for fragmentation can, in fact, go to zero, if less than a critical percentage saturation is achieved for a given initial condition and final quench temperature. This would occur, for example, in Fig. [22], if starting at 800°C and 1% saturation the metal was quenched to 300°C. No supersaturation would occur and in fact, a negative latent pressure would exist so that there would be no driving force for fragmentation.

5.2 The Effect of Reduced Pressure

The effect of initiating the quench from a reduced system pressure may be seen readily from inspection of Fig. [21]. If the initial pressure at which the molten metal

is heated is, say, 10^{-5} atmospheres and the temperature is 500°C -- point A', then quenching the drop to say 420°C in water would result in state B' if the driving force for fragmentation is considered to be the latent pressure. It is evident that the driving force for fragmentation is substantially decreased for lowered initial pressures.

5.3 The Effect of the Initial Drop Temperature

A drop initially heated to a higher temperature A3 of Fig. [21], would have a greater concentration of gas when it reached saturation. Such a drop when cooled rapidly to the same temperature B3 would have a greater latent pressure driving force for gas dissolution.

5.4 Experimental Plan

It was decided to investigate the validity of the Epstein hypothesis experimentally by utilizing two different heating modes prior to dropping the molten tin drops into cold water. One mode involved heating the tin in an air atmosphere, the other involved heating in a moderate, 10^{-5} atm, vacuum.

A nitrogen atmosphere was used initially instead of a vacuum since nitrogen is insoluble in tin, but extensive fragmentation resulted and it was suspected that impurities in the nitrogen might account for this under the gas evolution theory. To avoid this we went to heating in a

vacuum and monitored the outgassing as reported in Section 5.5. If the cause of fragmentation was indeed gas dissolution, one would expect a much smaller driving force for fragmentation at reduced pressure as indicated in Section 5.2 and therefore, either no fragmentation or a considerably reduced violence of interaction. If, on the other hand, gas dissolution was not the primary cause of fragmentation one would expect no real difference between the experiments conducted with heating in air and those conducted with heating in a vacuum.

5.5 The Experimental Apparatus and Procedure

The apparatus used is shown in Fig. [23]. It is split up into two chambers, the upper one which may be evacuated with a vacuum pump and the lower one which contains distilled water at room temperature and a pressure transducer. The two chambers are separated by a trapdoor valve.

The tin is heated inside the crucible which is placed in the upper chamber. The crucible is closed at the bottom by a shutter plate which may be rotated to let the tin fall through. This is done by turning a handle attached to a long rod which passes through a hollow rod welded to the crucible. This long rod is connected at its other end to the shutter plate.

After heating, the crucible is lowered to a position just above the water surface, and the molten tin is dropped

into the water. The pressure signals picked up by the transducer are recorded on a twenty millisecond time span on magnetic tape that can be played back on an oscilloscope. The recorder is triggered through a delayed trigger transfer from an oscilloscope which is itself triggered by the entry of the drop into the water. The temperature of the tin sample is measured with a rhodium-platinum thermocouple attached to the crucible. At lowered pressures, the system pressure is measured by means of a thermocouple pressure gauge.

Two gram charges of tin were melted by heating and then dropped into water. With the water temperature maintained constant at 2.15°C , the tin initial temperature was varied and pressure recordings were made of the interaction event. The peak "Big Bang" pressure which coincides with the instant of fragmentation (Bjornard [15]) are shown in Fig. [24].

The above procedure describes that followed for the experiments in which heating was done in air. In these cases the trap door valve connecting the two chambers was kept open.

For the experiments conducted at vacuum conditions, the lower chamber is initially filled with nitrogen under a slight pressure and the trapdoor valve connecting the two chambers is kept closed. The upper chamber is then evacuated. The pressure-time curve of the vacuum chamber without heating

the tin was first determined during evacuation over a period of three days, so that a steady state was reached. Then using this curve as a base, during the course of the experiments, the upper chamber was evacuated and the heating started only after the pressure-time curve of the experiment followed the steady state slope of the reference curve. The tin was heated to a temperature just above that of dropping and maintained at that level while the pressure was initially observed to rise, but would recover again as indicated by the dotted line in Fig. (25) and the pressure time curve would be parallel to the extrapolated curve of the chamber with the unheated tin or would coincide with it, at which point the heating was stopped. The average heating time was 35 minutes. It was thus ensured that outgassing had been accomplished as far as possible.

After evacuating the upper chamber to a pressure of 8 microns of mercury, or 10^{-5} atm, and heating the tin the sequence of steps for dropping the heated tin starts. The solenoid valve is opened to flood the vacuum chamber with nitrogen from the lower chamber. This equalizes the pressures across the trapdoor valve which falls open. The crucible is immediately lowered to just above the water surface and the tin is dropped into the pool.

5.6 Experimental Results

The results for both heating modes was extensive fragmentation. The quantitative data (the peak pressures

measured) are indicated on Fig. (24). It is evident that any differences between the behavior for the atmospheric and vacuum heating modes are well within the scatter of data.

5.7 Discussion of the Results

Although all gases cannot be expected to follow Sievert's Law, in general the solubility of gases in molten metals increases with increasing pressure as well as with increasing temperature. Thus the driving force for fragmentation should decrease substantially with decreasing system pressure at constant initial temperature as shown in Fig. (21). One would therefore expect either no fragmentation at all or a significantly reduced violence of interaction for the experiments conducted heating the tin under a vacuum. However, the experimental data shown in Fig. (24) indicates no such effect. Indeed there seems to be no real difference between the results from the two different heating modes. This comparative result is a key finding which argues against gas release as the operating mechanism for the observed fragmentation. As a point of interest the latent pressures were calculated for 500°C molten tin produced by both heating modes and dropped into 21.5°C water. This calculation requires identification of a soluble gas and a final quench temperature. Oxygen was selected based on the available experimental data which indicates it is the most soluble gas in tin. The solidification temperature of tin, 232°C, was

taken as the quench temperature. With these conditions and the further assumptions that the oxygen-tin system follows Sievert's law and that no oxide forms, the latent pressures were calculated as shown in Appendix 3. Pressures of 1143 atmospheres and 0.01 atmospheres were obtained for the modes of heating in air and heating in vacuum, respectively. These pressures show a convincing difference and the latent pressure for heating in a vacuum seems too low compared to values which might be imagined as necessary to promote fragmentation.

In addition to the direct comparison of pressure result for every tin temperature from the two heating modes, the trend of both sets of results with initial tin temperature is also a key finding. The effect of changing the initial tin drop temperature is to start at a point such as A3 Fig. (21), and to cool to the final quench temperature already selected as the melting point of tin. The effect of drop temperature increase should then be to increase the driving force for fragmentation. The experiments should, therefore, show a monotonic increase in fragmentation violence with initial tin temperature; however, Fig. (24) shows a decrease in the peak pressure magnitude at somewhat higher temperatures for both heating modes.

It is possible that both these findings are due to the presence of a gas in the tin which does not obey Sievert's law. Such a possibility cannot be ruled out directly. However, in order to satisfy the observed trend in the peak

pressurs with initial drop temperatures, one would have to have a gas that would dissolve in molten metal and whose solubility would have to decrease with temperature in the observed temperature range. In addition to showing the independence of the results with heating mode the solubility of such a candidate gas would also have to be totally unaffected by the system pressure.

5.8 Slab Experiments

A final set of experiments was performed by dropping tin on slabs to perform the quench in place of water. If the gas hypothesis were the governing mechanism, the quench material would have been expected to have no effect on results. Only the tin cooling rate would have been important. In the first instance hot molten tin was dropped on a room temperature slab of aluminum. A simple calculation (see Appendix 2) indicates that in this situation, the tin cools considerably faster than when it is dropped into cold water. No interaction or fragmentation was observed. The experiment was repeated using a slab of fire-brick; here, the instantaneous cooling rate of the tin is expected to be close to that when water is used. No interaction was observed in this case either. Under the gas evolution hypothesis, one would expect fragmentation to have occurred in both cases since the cooling rates are equal to or greater than with water and in all water quenched cases fragmentation was observed.

Thus it does seem that not only is the gas evolution

hypothesis not applicable but also that a key factor for free contact fragmentation is that a liquid-liquid system is required. This has been suggested by several others, e.g. Fauske (12).

5.9 Conclusion

Considering the arguments just put forward it seems probable that violent release of dissolved gas from within the drop is not responsible for free contact fragmentation, at least for the drop experiments of tin. Further a liquid-liquid system is required for this mode of fragmentation.

CHAPTER 6

CONCLUDING REMARKS

6.1 General Conclusions

1. Based on the discussion in Chapter 5, it seems reasonable to say that the cause of fragmentation of liquid metals in water is not the evolution of dissolved gas from within the metal drop.
2. The slab experiments of Chapter 5 suggest that a liquid-liquid system is necessary for fragmentation to occur.
3. Based on the experiments in Chapter 2, a measure of the intensity of the fragmentation interaction is the peak 'Big Bang' pressure, experimentally recorded by a transducer.
4. Based on Chapter 3 it seems likely that the cause of fragmentation is nothing as simple as the vapor pressure generated around the drop by simple vaporization overcoming the surface tension of the metal drop.
5. While homogeneous nucleation of the liquid coolant can account for the high vapor pressures that might be thus produced and thereby cause fragmentation at somewhat higher initial drop temperatures, conditions which yield interface temperatures below the homogeneous nucleation temperatures. However the possibility of spontaneous nucleation as a mechanism is still open.

6. Based on the rough calculation in Chapter 4 it would seem that there is a high probability of a high velocity coolant jet striking the drop. This hypothesis would seem worth investigating.
7. All hypotheses advanced to date predict a monotonically increasing amount of fragmentation with initial metal temperature; none will therefore be satisfactory at somewhat higher temperatures where the degree of fragmentation is seen to decrease.

6.2 Recommendations for Future Work

1. The theory of coolant entrapment by jet penetration would seem worth investigating analytically, perhaps by refining the analysis of Buchanan et al to include the effects of the initial temperature of the drop and coolant.
2. Experiments could be run similar to those of Bjornard [15] but using very smooth solid spheres instead of molten metal and picking up the pressure behavior with a transducer to see if spontaneous nucleation would then occur in the absence of preferred nucleation sites, and thus yield pressure responses similar to those of the molten metal experiments rather than those of the solid drops reported in Chapter 2.

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APPENDIX 1

COMPARISON OF THE INTERFACE TEMPERATURE
WITH THE HOMOGENEOUS NUCLEATION TEMPERATURE

The homogeneous nucleation temperature of water at 1 atm. pressure is 314°C. In order to test the possibility of the water being superheated to this degree, the following calculation was performed. As the hot molten metal and cold water come into contact, the interface temperature T_1 assumes a value instantaneously given by:

$$T_1 = T_h + (T_h - T_c) (y/1 + y) \quad (21)$$

where

$$y = ((\rho k c)_h / (\rho k c)_c)^{1/2} \quad (22)$$

since at the first instant of contact, the metal and the water each present semi-infinite bodies to each other and the thermal boundary layer has not yet penetrated into either medium. This interface temperature may be expected to decrease as time progresses, so that the value given by eq. (21) may be regarded as an upper limit. In many cases, this value T_1 was substantially smaller than the homogeneous nucleation threshold. It would thus appear that reaching the homogeneous nucleation threshold by the coolant is not necessary for fragmentation to occur. The theory of spontaneous nucleation, however, is still open as a possibility.

In Figures (A1) to (A5), adapted from Cho et al [7] and others, the initial interface temperatures have been calculated from the initial drop and water temperatures by equations (21) and (22) and plotted against the extent of fragmentation, to show the effect of the important parameters. Although the minimum film boiling point correlation by Henry may not be applicable to liquid-liquid systems, it has been included in some of the figures.

A. Coolant Water

Effect of Interface Temperature (Fig. (A1) based on Ref. [7]

An increase of fragmentation with rising interface temperature is noted initially but there is a fall at higher temperatures.

Effect of Subcooling of Water (Fig. (A2)) based on Ref. [27]

Generally, with greater subcooling of water all else including interface temperature being equal, fragmentation was found to increase.

Effect of the Weber Number (Fig. (A3)) based on Ref. [7]

Fragmentation was found to increase with an increase in the number based on the entrance velocity of the drop $N_{We} = \rho_l v^2 D / \sigma$.

Effect of Dwell Time (Fig. (A4)) based on Ref. [28]

The dwell time is defined as the time between the entry

of the hot drop into the coolant and the explosive interaction. Fragmentation is seen to increase with dwell time.

Effect of Oxidation

McCracken [19] points out that results depend markedly on the amount of oxidation of the hot metal sample. If, before each test, the liquid metal surface was not scraped clean, inconsistent results were obtained. The presence of oxide on the surface is found to restrict fragmentation. This could account for the decrease in fragmentation observed at elevated temperatures.

The interface temperature (T_i) is dependent on the value of ($\rho k c$) of the molten drop. This value is seen to be lower for the oxide than for the metal. This might hinder fragmentation at higher molten drop temperatures when more oxide forms, by lowering the interface temperature.

Thus: (the values below are averaged over the range of working temperatures)

<u>Tin</u>		<u>Tin Oxide</u>	
$\rho k c$	.034	$\rho k c$	.026
<u>Zinc</u>		<u>Zinc Oxide</u>	
$\rho k c$	0.111	$\rho k c$	.025-.008

where $\rho k c$ is in $(\text{g/cm}^3)(\text{cal/sec/cm}^2\text{C})(\text{cal/g}^\circ\text{C})$.

Note: Besides the work of the authors mentioned here and earlier, there have been many other experimental contributions,

most of them quantitative. Experiments of the sort conducted at M.I.T. and by Cho et al [7] may be found in [31] and [32].

B. Coolant -- Liquid Sodium -- References [29][30]

Experiments using liquid sodium as the coolant are largely qualitative in nature.

Effect of Interface Temperature (Fig. (A5)) based on Ref. [29]

Particle size of the uranium oxide, both ejected and left behind is shown in Fig. (A5).

Qualitative Results Indicate:

- a) at a fixed sodium pool temperature, fragmentation is enhanced by an increase in initial drop temperature.
- b) at a fixed drop temperature, fragmentation is increased by increasing the sodium pool temperature.

Conclusions

1) A lot of quantitative data is from the work of Cho, Gunther and Armstrong [7]. They show a photograph of an experiment conducted twice under "identical" conditions of sample size, temperatures, entrance velocity, using tin and water. In one case, the fragmentation is slight; in the other it is extensive. This could mean that a factor, hitherto not considered, is involved in fragmentation. This may be the oxide formation suggested by McCracken.

2) Fragmentation is seen to occur well before the homogeneous nucleation temperature is reached at the interface. It, therefore, can not be solely responsible for fragmentation. It is possible, however, that heterogeneous nucleation for imperfect wetting (contact angle non-zero) occurs before homogeneous nucleation does, as proposed by Fauske.

APPENDIX 2
COOLING OF THE MOLTEN TIN WHEN
DROPPED ONTO LARGE SLABS

As indicated in Chapter 5, a small quantity of hot molten tin was dropped in turn onto large slabs of aluminum and fire-brick at room temperature. In neither case was any fragmentation observed. The of the calculations below is to see how fast the molten tin cools down and freezes when dropped onto the slabs, and to check that the slab itself does not vary significantly in temperature during the process.

Aluminum Slab-Cooling Rate

The instantaneous temperature predicted by eq. (21) - Appendix 1, is lower than the freezing point of tin, insce,

$$(\rho kc)_{Al} > (\rho kc)_{H_2O}$$

and the cooling rate may be expected to be more rapid. In order to check that the aluminum slab did not itself cool down substantially before the tin froze the following calculation was made, where the dimensions are shown in Figure A1.

If a phase change occurs, eq. (21) is not applicable. For a rough estimate it was assumed that the interface temperature initially assumes a value equal to the freezing point of the tin; the freezing front then advances in the

tin at a rate roughly:

$$w = \left(\frac{2k\theta_s}{\rho h_{if}} \right)^{1/2} \sqrt{t} \quad \text{where } \theta_s = T_{tin} - T_{mpt}$$

The time for all of the tin to freeze, t_1 , when $w = h$ is:

$$t_1 = \left[\frac{\rho h_{if}}{2k\theta_s} \right]_{Sn} h^2$$

A boundary layer grows in the aluminum at a rate proportional to $\sqrt{a_{tAl}} \sqrt{t}$. The time for the boundary layer to reach the other end of the slab is:

$$t_2 \approx H^2 / (a_t)_{Al}$$

$$t_1/t_2 \approx \left(\frac{h_{if}\rho}{2k\theta_s} \right)_{Sn} (a_t)_{Al} h^2 / H^2$$

Substituting for these properties,

$$t_1/t_2 \approx 119 \quad 6 (h/H)^2$$

since

$$(h/H) \ll 1/10$$

$$(t_1/t_2) \ll 1$$

so that by the time the tin has frozen, the aluminum has not cooled down to any extent.

Fire-Brick Slab-Cooling Rate of the Tin

The thermal properties of fire-brick are:

$$\rho = 145 \text{ lb/ft}^3$$

$$k = 0.79 \text{ BTU/hr. ft } ^\circ\text{F}$$

$$c = 0.23 \text{ BTU/lb } ^\circ\text{F}$$

$$\rho k c = 26.3 \text{ with the above units}$$

which is close to that of water in that the product $\rho k c$ is 21.2 for water. The instantaneous temperature on contact, given by eq. (21) may thus be expected to be close to that when the coolant is water.

APPENDIX 3

CALCULATION OF LATENT PRESSURE FOR TIN-OXYGEN

The reported solubility of oxygen in tin [25] was used to generate the graph Fig. A2 by drawing straight lines, on a log-log graph, of slope = $1/2$, through individual points, obtained from the solubility characteristics from Ref. [25] at various temperatures and 1 atm pressure. As explained in Chapter 5, Sievert's law is used even though it might not be accurate, to obtain rough estimates of latent pressure.

From the graph, and since Sievert's law is assumed to apply,

$$C_O = AP_O^{1/2} e^{-\Delta h_O/RT}$$

At 1 atm pressure,

$$\text{at } T = 873^\circ\text{K} \quad C_O = 10^{-2.5} \text{ weight percent}$$

$$\text{at } T = 673^\circ\text{K} \quad C_O = 10^{-3.45} \text{ weight percent}$$

$$\text{Dividing: } e^{-\Delta h_O/RT} (1/873 - 1/673) = 8.93$$

$$\text{This gives: } \Delta h_O/R = 6300^\circ\text{K}$$

Consider a tin droplet initially at 500°C dropped into water at room temperature. If the temperature used as a base to calculate latent pressures is taken to be the freezing point of tin, 232°C , the latent pressure may be readily

calculated:

$$C_O = AP_O^{1/2} e^{-h_O/RT}$$

Thus at 500 C or $T_1 = 773^\circ K$

$$(C_O)_{500} = AP_{O1}^{1/2} e^{-6300/T_1}$$

and at the reference temperature $T_2 = 505^\circ K$

$$(C_O)_{282} = AP_{O2}^{1/2} e^{-6300/T_2}$$

where $T_1 = 773^\circ K$ and $T_2 = 505^\circ K$

Dividing:

$$\left(\frac{P_{O2}}{P_{O1}}\right)^{1/2} = e^{-6300\left(\frac{1}{773} - \frac{1}{505}\right)} = 75.6$$

Since $(C_O)_{500}$ concentration is retained instantaneously at a temperature $232^\circ C$ and the latent pressure may be calculated by equating $(C_O)_{500} = (C_O)_{232}$.

For the "vacuum" experiments the initial pressure was 8 microns of mercury pressure = 1.07×10^{-5} atmospheres. If P_{O1} , the partial pressure of the oxygen present, is taken to be 0.2 times this pressure, then,

$$P_{O1} = 0.2 \times 1.07 \times 10^{-5} = 2.1 \times 10^{-6} \text{ atm.}$$

$$\text{Then } P_{O2} = (75.6)^2 (2.1 \times 10^{-6}) = 0.01 \text{ atm}$$

And the latent pressure is only 0.01 atm.

By contrast for the atmosphere experiments,

$$P_{O1} = 0.2 \text{ atm}$$

$$P_{O_2} = (75.6)^2 (0.2) = 1143 \text{ atmospheres}$$

and the latent pressure is 1143 atmospheres!

APPENDIX 4

EXPERIMENTAL CHECK OF EQUATION (1)

In order to verify the validity of eq. (1) which predicts that the pressure in the liquid decreases inversely as the distance from the drop, experiments were run dropping molten tin into water and using two pressure transducers in the water simultaneously, at different distances from the drop, with each transducer connected to a separate signal recorder, as shown in Fig. (A8). This was done to eliminate the problem of data scatter from interfering with the comparison between the readings of the two transducers. The results are plotted on Figure (A9) and show that while the data points fall evenly on both sides of the line represented eq. (1) there is too much scatter to permit continued use of the formula. The box or line around a data point indicates the uncertainty in measurement.

$\bar{z}_1$ = distance of transducer 1 from the drop

$\bar{z}_2$ = distance of transducer 2 from the drop

APPENDIX 5

A MODEL FOR THE HEAT TRANSFER FROM THE DROP

A model is proposed for the fluid flow and heat transfer phenomena occurring between the molten metal drop and the coolant.

In a frame of reference fixed to the drop, the coolant rushes upwards towards it, and flows past it as shown in Figure (A10). It is assumed that the drop carries with it some vapor all around it, and that the drop does not deform appreciably.

The following assumptions are made:

- a) During the time interval of interest, the sphere surface temperature remains constant.
- b) All the heat conducted from the hot sphere to the vapor-film liquid interface goes into evaporation of the liquid, i.e. none is conducted into the liquid.
- c) Pressure across the film is uniform but varies along the periphery of the drop.
- d) The liquid vapor interface is at saturation temperature T_{sat} corresponding to the film pressure.
- e) The temperature variation across the film is linear.
- f) In the coolant liquid, the flow is almost inviscid and may be regarded as potential.
- g) Separation occurs at the rear end of the drop. The pressure at this point is assumed to be atmospheric.

Then for the control volume shown, the following equations may be written:

$$q'' = K_v (T_w - T_{sat}) \delta \quad (23)$$

where q'' is the heat flux from the drop.

Mass rate of evaporation into control volume,

$$\dot{m}_1 = q'' \pi 2 (R_1 + \delta)^2 \sin \theta d\theta / h_{fg} \quad (24)$$

where R_1 is the drop radius.

Mass rate of efflux of mass out of control volume due to flow of vapor past sphere,

$$\dot{m}_2 = \rho \bar{V} \int_0^\pi 2\pi (R_1 + \delta/2) \sin \theta \delta + \frac{\partial}{\partial \theta} [\rho \bar{V} \delta 2\pi (R_1 + \delta/2) \sin \theta] d\theta \quad (25)$$

where $\bar{V}$ is the average vapor flow velocity in the film and ρ is the density of the vapor.

Rate of influx of mass into control volume due to inflow of vapor

$$\dot{m}_3 = \rho \bar{V} 2\pi (R_1 + \delta/2) \sin \theta \quad (26)$$

Because of these mass fluxes, the mass inside the control volume will vary with time as,

$$\frac{\partial m}{\partial t}_{c.v.} = \frac{\partial}{\partial t} [\rho 2\pi \delta (R_1 + \delta/2)^2 \sin \theta] d\theta \quad (27)$$

Combining equations (23 - 27) into an equation of mass conservation:

$$\frac{K_v (T_w - T_{sat})}{\delta h_{fg}} = \frac{2\pi (R_1 + \delta)^2 \sin \theta}{\partial \theta} \left[\rho \bar{v} - \frac{\partial}{\partial \theta} \left(\rho \bar{v} \frac{2\pi (R_1 + \delta/2)}{\sin \theta} \right) \right] = \frac{\partial}{\partial t} \left[\rho 2\pi \delta (R_1 + \delta/2)^2 \sin \theta \right] \quad (28)$$

for the vapor we may write,

$$P = \rho R (T_w + T_{sat})^{1/2} \quad (29)$$

where R is the perfect gas constant, assuming it to act as a perfect gas. As the saturation curve for the coolant relates the saturation pressure and temperature,

$$P = P(T_{sat}) \quad (30)$$

The average velocity in the film may be calculated by assuming a velocity profile, e.g. parabolic as in Couette flow between parallel plates of height δ , driven by a pressure gradient given by eq. (33).

The boundary conditions would be:

$$\begin{aligned} v_\theta &= 0 \text{ at surface of sphere } r = R_1 \\ v_\theta &= 3/2 V_\infty \sin \theta \text{ at } r = (R_1 + \delta) \end{aligned} \quad (31)$$

assuming no slip at the liquid-vapor interface and potential flow past a sphere in the liquid, with a free stream velocity of V_∞ .

The latent heat may be related to T_{sat} at the cold liquid interface, for the particular liquid, by,

$$h_{fg} = f(T_{sat}) \quad (32)$$

Also there is a pressure gradient in the liquid coolant which is impressed on the film. This pressure gradient may be evaluated by the application of Benoulli's theorem as:

$$\partial P / \partial \theta = -\rho_1 V \partial V / \partial \theta \quad (33)$$

where

$$V = 3/2 V_\infty \sin \theta \quad (34)$$

the velocity in the liquid.

An initial film thickness may be assumed smaller than the steady estimate of 4.2. Also, pressure at the position corresponding to $\theta = \pi/2$ in the film is assumed to be atmospheric. Equations (28-34) may now be solved using the method of finite differences to predict the film behavior and the heat transfer from the drop.

APPENDIX 6
EXPERIMENTAL DATA

✓

EXPERIMENTS WITH SINGLE SOLID SPHERES - SERIES #

Drop Material: Steel CR52-100, a chrome steel

Drop Radius: 0.4 cm

Temperature of Water: 23°C

Transducer Depth: 6 1/2" (unless otherwise stated)

Drop Height: 2 11/16"

Sensitivity Setting on Recorder: 4 mv/division

<u>Run No.</u>	<u>Drop Temperature</u> <u>(°C)</u>	<u>Recorder Delay</u> <u>(msec)</u>	<u>Peak Pressure at Transducer</u> <u>(psi)</u>	<u>Comment</u>
1	540	75	0.19	signal has
2	540	60	0.27	died down
3	540	45	0.68	signal has
4	340	45	0.97	died down
5	390	45	0.55	
6	340	45	0.12	
7	340	45	0.07	
8	390	45	0.41	
9	440	45	0.20	
10	440	45	0.51	

<u>Run No.</u>	<u>Drop Temperature</u> (°C)	<u>Recorder Delay</u> (msec)	<u>Peak Pressure at Transducer</u> (psi)	<u>Comment</u>
11	515	45	1.20	
12	590	45	0.04	
13	540	55	1.00	
14	590	55	0.07	
15	620	50	0.07	
16	620	60	0.07	
17	620	60	0.07	
18	590	60	0.06	signal has ∞
19	620	75	0.034	died down ∞
20	440	48	0.68	(transducer depth 2 1/2")

8 SOLID SPHERES INTO WATER SERIES B

Drop Material: Steel

Drop Radius: 0.2 cm

Transducer Depth: 3 3/4"

Temperature of Water: 23.5°C

Dropping Height: 2 11/16"

<u>Run No.</u>	<u>Drop Temperature</u> <u>(°C)</u>	<u>Recorder Delay</u> <u>(msec)</u>	<u>Peak Pressure</u> <u>(psi)</u>	<u>Heating Time</u> <u>(minutes)</u>
B1	390	45	0.61	0
B2	440	42	0.11	0
B3	440	45	0.17	0
B4	490	40	-	0
B5	490	50	0.14	0
B6	540	45	0.34	0
B7	590	42	-	0
B8	590	52	0.22	0
B9	640	48.5	-	0
B10	640	53	0.31	0
B11	390	45	-	15
B12	390	50	0.19	20

<u>Run No.</u>	<u>Drop Temperature</u> <u>(°C)</u>	<u>Recorder Delay</u> <u>(msec)</u>	<u>Peak Pressure</u> <u>(psi)</u>	<u>Heating Time</u> <u>(minutes)</u>
B13	390	50	0.15	0
B14	440	45	0.29	20
B15	440	55	0.34	20
B16	440	67.5	0.11	20
B17	440	55	0.68	0
B18	490	50	1.02	0
B19	490	45	0.51	0
B20	540	45	0.15	0
B21	590	45	0.15	0
B22	640	48.5	0.34	0
B23	490	45	0.08	45
B24	540	50	0.20	20
B25	590	50	0.07	20

EXPERIMENTAL CHECK OF EQUATION (1) - SERIES D

Drop Material: Tin

Drop Temperature: Variable

Water Temperature: 23°C

Transducers Distance: Variable ($\bar{z}_1, \bar{z}_2$ measured, $\pm z_1 z_2$ measurement uncertainty)

Sensitivity of Recorders: 4mVolts/division

Recorder Delay: 45 msec

Drop Mass: Variable

Note: P_1/P_2 was calculated directly from the two recorded signals, rather than from the pressures shown in the columns, in the interests of accuracy.

Run No.	Tin Temp. (°C)	$\bar{z}_1$ (in)	$\pm z_1$ (in)	$\bar{z}_2$ (in)	$\pm z_2$ (in)	z_2/z_1	Peak Pressure at transducer 1 P_1 (psi)	Peak Pressure at at Xaucer 2 P_2 (psi)	P_1/P_2
D1(a)	440	$3 \frac{1}{16}$	$\pm \frac{1}{16}$	7"	$\pm \frac{1}{2}$	2.33	0.61	0.17	3.6
(b)	490						0.17	0.07	2.5
(c)	340						0.11	0.02-0.03	6.5-3.3
(d)	390						0.11	0.02-0.03	6.5-3.3
(e)	540						0.65	0.20	3.2
D2(a)	340						0.10-0.12	0.03-0.04	2.4-3.5
(b)	340						0.94	0.09	11.0

Run No.	Tin Temp. (°C)	$\bar{z}_1$ (in)	$\pm z_1$ (in)	$\bar{z}_2$ (in)	$\pm z_2$ (in)	$\bar{z}_2/\bar{z}_1$	Peak Pressure at Transducer 1 P_1 (psi)	Peak Pressure at Transducer 2 P_2 (psi)	P_1/P_2
D2 (c)	340	$1 \frac{1}{2}$ "	0	$3 \frac{1}{16}$ "	$\pm \frac{1}{4}$ "	2.08	0.85	0.10	8.3
(d)	340						0.27	0.94	0.3
(e)	340						0.23	0.07	3.4
(f)	340						0.51	0.09	6.0
D3 (a)							0.14	0.03	4.0
(b)							0.10	0.05	2.0
(c)							0.37	0.0 - 0.07	7.3-5.5
(d)	440	3"	$\pm \frac{1}{16}$ "	$4 \frac{1}{2}$ "	$\pm \frac{1}{4}$ "	1.50	0.11	0.07	1.6
(e)							0.20	0.14	1.5
(f)							0.26-0.34	0.17	1.5-2.0
D4 (a)							0.10-0.14	0.03	3.0-4.0
(b)							0.13	0.04	3.0
(c)	440	2"	$\pm \frac{1}{16}$ "	6"	$\pm \frac{1}{2}$ "	3.00	0.10	0.03	3.0
(d)							0.22	0.34	0.65
D5 (a)	420						0.03	0.05	1.5
(b)	440						0.41	0.41	1.0

Run No.	Tin Temp. (°C)	$\bar{z}_1$ (in)	$\pm z_1$ (in)	$\bar{z}_2$ (in)	$\pm z_2$ (in)	$\bar{z}_2/\bar{z}_1$	Peak Pressure at Transducer 1 P_1 (psi)	Peak Pressure at Transducer 2 P_2 (psi)	P_1/P_2
D5 (c)	440	$4 \frac{1}{4}$ "	0	$4 \frac{1}{4}$ "	0	1.00	0.41		
(d)	470						0.09	0.09	1.3
(e)	440						0.04	0.03	1.0
(f)	440						0.03	0.03	1.0
D6 (a)							0.10	0.03	3.0
(b)							0.05	0.04	1.2
(c)	440	$1 \frac{9}{16}$ "	0	$3 \frac{2}{16}$ "	0	2.00	0.12	0.06	2.0
(d)							0.09	0.03	2.5

VACUUM EXPERIMENTS WITH MOLTEN TIN - CHAPTER 5 - SERIES SG

Drop Material: Molten Tin

Dropping Height: 1"

Drop Mass: 2 g (Radius 0.41 cm)

Sensitivity of Recorder: 4 mVolts/Division

Temperature of Water: 21.5°C

Pressure in Heating Chamber: 8×10^{-6} m. of
mercury = 1.07×10^{-5} atmospheres

Transducer Depth: 4"

<u>Run No.</u>	<u>Tin Temperature</u> <u>(°C)</u>	<u>Recorder Delay</u> <u>(msec)</u>	<u>Heating Time</u> <u>(minutes)</u>	<u>Peak Pressure</u> <u>at Transducer</u> <u>(psi)</u>	<u>Comments</u>
SG 1	390	16	48	0.22	
SG 2	415	20	25	1.53	
SG 3	440	25	39	1.02	
SG 4	490	22.5	17	0.07	
SG 5	490	22.5	28	0.10	
SG 8	440	22.5	15	0.31	
SG 9	540	22.5	15	0.39	
SG 10	440	22.5	35	0.14	
SG 11	580	25	30	1.53	
SG 12	640	28	50	0.10	

ATMOSPHERE EXPERIMENTS WITH MOLTEN TIN - CHAPTER 5 - SERIES SGA

Drop Material: Tin

Drop Mass: 2 g. (Radius 0.41 cm)

Water Temperature: 21.5° C

Drop Temperature: Variable

Transducer Depth: 4 inches

Dropping Hight: 1 inch

Recorder Sensitivity: 4 mVolts/division

Heating Time: less than 1 minute

Pressure in Heating Chamber: Atmospheric

<u>Run No.</u>	<u>Tin Temperature</u> <u>(°C)</u>	<u>Recorder Delay</u> <u>(msec)</u>	<u>Peak Pressure</u> <u>at Transducer</u> <u>(psi)</u>	<u>Comments</u>
SGA 1	390	16	0.06	
SGA 2	390	16	0.10	
SGA 3	440	20	0.29	
SGA 4	440	20	0.18	
SGA 5	540	22.5	0.51	
SGA 6	540	22.5	1.11	
SGA 7	640	40	0.09	No Big Bang
SGA 8	640	60	0.10	No Big Bang

FIG. 1: RESULTS OF DROPPING VARIOUS MATERIALS IN WATER (REF 34)

FIG. 2: FRAGMENTATION OF MOLTEN TIN DROPPED INTO WATER
AT VARIOUS TEMPERATURES (REF. 7)

FIG. 3: EFFECT OF INITIAL DROP TEMPERATURE ON PEAK PRESSURE
 AT DROP (REF 16)

FIG. 4: EFFECT OF INITIAL DROP TEMPERATURE ON PEAK PRESSURE
AT TRANSDUCER (REF 16)

FIG. 5: EFFECT OF INITIAL DROP TEMPERATURE ON DWELL TIME (REF 16)

FIG. 6: TEMPERATURE INTERACTION ZONE FOR TIN AND WATER - FROM REF. (20)

FOR 12 GRAMS OF TIN DROPPED THROUGH 3 cm INTO
 -BOILED DISTILLED WATER o INDICATES AN INTERACTION
 x INDICATES NO INTERACTION

FIG. 7: APPARATUS FOR SOLID SPHERE EXPERIMENTS
FROM REF [16]

FIG. 8: ELECTRONICS CIRCUIT FOR RECORDING PRESSURE

FIG 10: EFFECT OF SOLID BALL TEMPERATURE ON PEAK PRESSURE

MATERIAL: 8 SOLID STEEL BALLS
DROP RADIUS: 0.2 cm
WATER TEMP.: 23.5°C
DROP TEMP.: VARIABLE
TRANSDUCER DEPTH: 3 3/4"

FIG. 11: EFFECT OF SOLID BALLS' TEMPERATURE ON PEAK
PRESSURE

TIN INTO WATER
 WATER TEMP: 20°C
 DROP TEMP: VARIABLE

FIG 12: INITIAL WEBER NUMBER MODEL - EFFECT OF TIN TEMPERATURE
 BASED ON REF [5]
 FOR $\delta_0 = 10^{-5}$ cm

FIG 13: INITIAL WEBER NUMBER MODEL - EFFECT OF WATER TEMPERATURE

BASED ON REF [5] $\delta_0 = 10^{-5}$ cm

FINAL WEBER NUMBER N_{We}

TIN DROPPED INTO WATER

<u>No.</u>	<u>Tin Temp.</u> (°C)	<u>Water Temp.</u> (°C)	<u>Final Diameter</u> D cm.	<u>Peak Pressure</u> P_{MAX} psi	<u>N_{We}</u>
1	400	22	0.0916	24.38	2398
2	500	4	0.0717	25.30	2055
3	350	22	0.1650	23.64	3988
4	300	4	0.1950	18.93	2230
5	500	22	0.0782	30.50	3340
6	600	22	0.2150	52.70	22089

BISMUTH DROPPED INTO WATER

<u>No.</u>	<u>Bismuth Temp.</u> (°C)	<u>Water Temp.</u> (°C)	<u>Final Diameter</u> D cm.	<u>Peak Pressure</u> P_{MAX} psi	<u>N_{We}</u>
7	500	22	0.0456	26.25	1970
8	500	50	0.1469	40.34	14042
9	350	50	0.1000	28.23	5042
10	350	4	0.0500	18.01	617

FIG. 14: FINAL WEBER NUMBER MODEL RESULTS - BASED ON REF [5]

FIG 15: TIME INTEGRATED PRESSURE - EFFECT OF TIN TEMPERATURE
BASED ON REF. [5]

DROP MATERIAL: TIN
 DROP TEMP: 500°C
 WATER TEMP: VARIABLE

FIG 16: TIME INTEGRATED PRESSURE EFFECT OF WATER TEMPERATURE
 BASED ON REF [5]

WATER TEMP: 20°C
TIN TEMP: VARIABLE

FIG. 17: EFFECT OF TIN TEMPERATURE ON TIME INTEGRATED PRESSURE BASED ON REF. [15]

TIN INTO WATER
 TEMP. OF TIN: 500°C
 TEMP. OF WATER: VARIABLE

FIG. 18: EFFECT OF WATER TEMPERATURE ON TIME INTEGRATED PRESSURE BASED ON REF [15]

FIG 19: DROP FALLING THROUGH COOLANT

FIG. 20: TIME REQUIRED TO DEFORM THE DROP τ

FIG. 21: ILLUSTRATION OF SIEVERT'S LAW AND CALCULATION OF LATENT PRESSURE

FIG. 22: EFFECT OF PERCENTAGE SATURATION OF METAL ON LATENT PRESSURE

FIG. 23: VACUUM HEATING APPARATUS

FIG. 24: RESULTS OF VACUUM HEATING AND AIR HEATING EXPERIMENTS

FIG. 25: PRESSURE HISTORY OF THE VACUUM CHAMBER

MOLTEN TIN DROPPED INTO 22°C WATER

DROP TEMP: VARIABLE

FIGURE A1: EFFECT OF INTERFACE TEMPERATURE
BASED ON REF. [7]

FIG. A3: EFFECT OF WEBER NUMBER

FIG. A4: EFFECT OF DWELL TIME
BASED ON REF. [28]

FIGURE A5: EFFECT OF INTERFACE TEMPERATURE - COOLANT SODIUM
BASED ON REF [29]

FIGURE A6: MOLTEN TINDROPPED ONTO A SOLID SLAB

FIGURE A7: SOLUBILITY OF OXYGEN IN MOLTEN TIN WITH PRESSURE, POSTULATING SIEVERT'S LAW

FIGURE A8: SET UP USED IN CHECKING EQUATION (1)

FIGURE A9: RESULTS OF EXPERIMENTS TO CHECK EQUATION (1)

FIG. A10 : DROP FALLING THROUGH COOLANT