APPLICATION OF SPONTANEOUS NUCLEATION THEORY TO
TIN-H₂O INTERACTIONS

by

M. Corradini
W.M. Rohsenow
N.E. Todreas

NOTICE
This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

January, 1977

Patent Cleared by Brookhaven Patent Office

Department of Nuclear Engineering
MASSACHUSETTS INSTITUTE OF TECHNOLOGY
77 Massachusetts Avenue
Cambridge, Massachusetts 02139

MASTER

Approved: _____

Neil E. Todreas

Neil E. Todreas
Principal Investigator

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

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)

W.F. Lenz, G. Shiralker and N. Todreas, Fuel Coolant Thermal Interaction Project UC 79P, COO-2781-1, Nov. 1975.

W.F. Lenz, G. Shiralker and N. Todreas, Fuel Coolant Thermal Interaction Project UC 79P, COO-2781-2, Feb. 1976.

G. Shiralker, W.F. Lenz and M. Corradini, Fuel Coolant Thermal Interaction Project UC 79P, COO-2781-3, April 1976.

G. Shiralker, W.F. Lenz and M. Corradini, Fuel Coolant Thermal Interaction Project UC 79P, COO-2781-5, Oct. 1976.

G. Shiralker, W.F. Lenz and M. Corradini, Fuel Coolant Thermal Interaction Project UC 79P, COO-2781-10, Dec. 1976.

Reports Issued under this Contract

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

Mujid S. Kazimi, "Theoretical Studies on Some Aspects of Molten Fuel-Coolant Thermal Interaction," 31-109-38-2831-1TR, MITNE-155, May 1973.

Charles E. Watson, "Transient Heat Transfer Induced Pressure Fluctuations in the Fuel-Coolant Interaction," 31-109-38-2831-2TR, MITNE-156, August 1973.

Trond A. Bjornard, "An Experimental Investigation of Acoustic Cavitation as a Fragmentation Mechanism of Molten Tin Droplets in Water," 31-109-38-2831-3TR, MITNE-163, May 1974.

Glen Bjorkquist, "An Experimental Investigation of the Fragmentation of Molten Metals in Water," 31-109-38-2831-4TR, June 1975.

Roland B. Knapp, "Thermal Stress Initiated Fracture as a Fragmentation Mechanism in the UO_2 -Sodium Fuel-Coolant Interaction," 31-109-38-2831-5TR, May 1975.

Michael Corradini, "Prediction of Minimum UO_2 Particle Size Based on Thermal Stress Initiated Fracture Model," COO-2781-4TR, August 1976.

M. Kazimi, C. Watson, D. Lanning, W. Rohsenow, N. Todreas, "Acoustic Cavitation as a Mechanism of Fragmentation of Hot Molten Droplets in Cool Liquids," COO-2781-6TR, November 1976.

Gautam S. Shiralkar, "An Investigation of the Fragmentation of Molten Metals Dropped into Cold Water," COO-2781-7TR, November 1976.

William F. Lenz, Jr. "Mixing Requirements for the Limiting Fuel-Coolant Interactions in Liquid Metal Fast Breeder Reactors," COO-2781-8TR, November 1976

M. Corradini, A.A. Sonin, N. Todreas, "A Proposed Heat Transfer Model for the Gas-Liquid Heat Transfer Effects Observed in the Stanford Research Institute Scaled Tests," COO-2781-9TR, December 1976.

M. Corradini, W.M. Rohsenow, N.E. Todreas, "Application of Spontaneous Nucleation Theory to Tin- H_2O Interactions," COO-2781-11TR, January 1977.

Reports Issued under this Contract

Papers and Summaries

M.S. Kazimi, N.E. Todreas, D.D. Lanning and W.M. Rohsenow, "A Criterion for Free-Contact Fragmentation of Hot Molten Materials in Coolants," Transactions of the American Nuclear Society, Vol. 5, No. 2, p. 835, November 1972.

M.S. Kazimi, N.E. Todreas, W.M. Rohsenow and D.D. Lanning, "A Theoretical Study of the Dynamic Growth of a Vapor Film Around a Hot Sphere in a Coolant," Fifth International Heat Transfer Conference, Tokyo, 1974.

T.A. Bjornard, W.M. Rohsenow and N.E. Todreas, "The Pressure Behavior Accompanying the Fragmentation of Tin in Water," Transactions of the American Nuclear Society, Vol. 19, pp. 247-248, 1974.

R. Knapp and N. Todreas, "Thermal Stress Initiated Fracture as a Fragmentation Mechanism in the UO_2 -Sodium Fuel-Coolant Interaction," Nuclear Engineering and Design 35, pp. 69-85, 1975.

"This report was prepared as an account of Government-sponsored work. Neither the United States, or the Energy Research and Development Administration nor any person acting on behalf of the Commission.

- A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or
- B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of, any information, apparatus, method, or process disclosed in this report.

As used in the above, 'person acting on behalf of the Commission' includes any employee or contractor of the Administration or employee of such contractor, to the extent that such employee or contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Administration or his employment with such contractor."

ACKNOWLEDGEMENTS

I would like to express my gratitude to Professor Neil Todreas for his concern, suggestions and active participation in the critique of this work.

Discussions with Professor Warren Rohsenow, and Dr. Robert Henry were also most helpful.

My deepest thanks goes to my wife Debi who helped in final preparation of the text and whose patient understanding was most valuable.

Sponsorship of this work was provided through the auspices of the Energy Research and Development Administration.

TABLE OF CONTENTS

List of Figures	11
List of Tables	v
Nomenclature	vi
1. INTRODUCTION	1
2. EXPERIMENTAL TIN-WATER DATA	4
2.1 Experimental Dependence on Temperature	5
2.2 Effect of Mass Size and Shape Interaction.	11
2.3 Dwell Time and Destabilization of Film Boiling	12
3. PROPOSED ANALYTICAL MODELS FOR FUEL COOLANT INTERACTIONS	32
3.1 Hydrodynamic Models	33
3.2 Violent Gas Release Model	34
3.3 Solid Shell Model	36
3.4 Molten Drop Models	36
3.4.1 Internal Acoustic Cavitation	37
3.4.2 Vapor Bubble Growth and Jet Model	38
3.4.3 Spontaneous Nucleation Model.	42
4. APPLICATION OF SPONTANEOUS NUCLEATION THEORY TO TIN-WATER EXPERIMENTS	66
4.1 Trigger Direct Liquid-Liquid Contact	66
4.1.1 Effect of Shape on Destabilization Temperature	67
4.1.2 Effect of Oxidation of Tin-Water Experiments	70
4.2 The Interaction as Depicted by the Spontaneous Nucleation Theory	72
5. CONCLUSION	94
APPENDIX A Film Collapse based on Notion of Helmholtz Instabilities	97
APPENDIX B Effect of Shape on Destabilization	102
Appendix C Possible Drop Deformation due to Weber No. Effects	105
APPENDIX D Effect of Oxide on Tin-Water Interaction	107
APPENDIX E Tin-Water Interaction as Predicted by Spontaneous Nucleation Theory	116
REFERENCES	124

LIST OF FIGURES

- Figure 2.1 Temperature Interaction Zone for 12 gm of Tin
- Figure 2.2 Sequence of Pictures of Debris as the TIZ is Crossed with MIT-UCLA data
- Figure 2.3 Temperature Interaction Zone data
- Figure 2.4 Typical Pressure Trace for Tin-H₂O Interaction
- Figure 2.5 Peak Pressure as a Function of Initial Fuel Temperature
- Figure 2.6 Fragmentation as a Function of Initial Fuel Temperature
- Figure 2.7 Peak Pressure as a Function of Initial Coolant Temperature
- Figure 2.8 Fragmentation as a Function of Initial Coolant Temperature
- Figure 2.9 Peak Pressure as a Function of Initial Tin Mass
- Figure 2.10 Dwell Time and Interaction Temperature as a Function of Initial Coolant Temperature
- Figure 2.11 Dwell Time and Interaction Temperature as a Function of Initial Fuel Temperature
- Figure 2.12 Comparison of Stevens-Witte Data to TIZ
- Figure 2.13 Peak Pressure as a Function of Fuel Temperature for Freon-Oil System
- Figure 2.14 Heat Transfer Coefficient for a Sphere in Water Inferred from Stevens-Witte [7].
- Figure 2.15 Heat Flux from a Hot Sphere in Water [7].
- Figure 3.1 Dissolved Gas Concentration as a Function of Gas Pressure

- Figure 3.2 Peak Pressure as a Function of Initial Fuel Temperature in a Vacuum
- Figure 3.3 Premixed Zone of Fuel in Coolant
- Figure 3.4 Pictorial View of Cold Drop Impinging on Hot Fuel Surface
- Figure 3.5 Temperature as a Function of Distance from Interface [11]
- Figure 3.6 Pictorial View of Film Boiling and Wetting and Capture
- Figure 3.7 Criterion of Wetting of Fuel (Oil) Surface by Coolant [11]
- Figure 3.8 Typical Heat Flux vs. Temperature Curve
- Figure 3.9 Two Different Views of Stable Film Boiling
- Figure 4.1 View of Growth of Tin Projection from Bulk of Tin Mass
- Figure 4.2 Model of Growing Projection Using a Sphere Projection
- Figure 4.3 Model of Growing Projection Using a Fixed Tin Projection
- Figure 4.4 Model of Formation of Tin-Oxide Shell on Molten Tin
- Figure 4.5 Criterion of Wetting of Fuel (Tin) Surface by Coolant
- Figure 4.6 Characteristic Depth (ℓ) as a Function of Initial Fuel Temperature for MIT Data
- Figure 4.7 Characteristic Depth (ℓ) as a Function of Initial Fuel Temperature for UCLA Data
- Figure 4.8 Characteristic Depth (ℓ) as a Function of Initial Fuel Temperature for MIT Data
- Figure 4.9 Qualitative Picture of Characteristic Depth as a Function of Initial Fuel Temperature

- Figure A.1 Pictorial View of Possible Helmholtz
Instability of Film Destabilization
Point
- Figure B.1 Approximate Model of Tin-Oxide Layer
Formation
- Figure E.1 Coolant Temperature as a Function
of Distance from Interface ($t = 10^{-7}$ sec) . .
- Figure E.2 Coolant Temperature as a Function of
Distance from Interface ($t = 10^{-6}$ sec) . . .
- Figure E.3 Coolant Temperature as a Function of
Distance from Interface ($t = 10^{-5}$ sec) . . .
- Figure E.4 Coolant Temperature as a Function of
Distance from Interface ($t = 10^{-4}$ sec) . . .

LIST OF TABLES

Table 2.1	Some Material Properties
Table 3.1	Homogeneous Nucleation Frequency as a Function of Temperature for Water
Table 3.2	Homogeneous Nucleation Frequency as a Function of Temperature for Freon-22
Table 3.3	Homogeneous Nucleation Frequency as a Function of Temperature for Sodium
Table 4.1	Tin Cooling Analysis
Table 4.2	Tin Cooling Analysis--Minimum Values for Cooling Projections
Table 4.3	Calculation of Characteristic Depth (ℓ) for Observed Pressure Pulse-MIT Data
Table 4.4	Calculation of Characteristic Depth (ℓ) for Observed Pressure Pulse-UCLA Data
Table 4.5	Calculation of Characteristic Depth (ℓ) for Observed Pressure Pulse-MIT Data
Table D.1	Properties of Tin-Oxide
Table D.2	Tin Cooling Analysis Based on Formation of Oxide Layer

NOMENCLATURE

A_{csec}	- cross sectional area of projecting fin
A_{es}	- cross sectional flow area in vapor space during film boiling
A_{sphere}	- surface area of fuel sphere
a	- radius of tin droplet when oxide layer is considered
c_{p_i}	- specific heat of ith material
C_g	- concentration of dissolved gas in metal
c	- acoustic velocity in coolant medium
d	- characteristic depth of coolant drop which participates in vaporization
D_D	- depth below surface coolant that pressure transducer is placed
D	- diameter of tin drop when assumed spherical
D_A	- diameter of initial vapor embryo after nucleation
D_B	- maximum diameter that vapor bubble can grow to
D'_b	- diameter of a vapor bubble nucleating at the limit near the critical temperature
E	- Young's Modulus
g	- gravity
ΔG	- change in Gibb's free energy due to bubble formation
h_i	- heat transfer coefficient i - c for film coefficient s for oxide coefficient
h	- Planck's Constant
h_{fg}	- heat of vaporization
ΔH	- change in enthalpy due to dissolving the gas in metal
J	- homogeneous nucleation frequency per unit volume per unit time

k	- Boltzman constant
k_i	- thermal conductivity of the i th material (c - coolant, f - fuel, SO - oxide)
K	- Bulk Modulus
L	- length of projecting fin
ℓ	- characteristic depth for pressure relief
m_v	- mass flow of vapor
$N_{\max}$	- maximum bubble population
N_{limit}	- maximum bubble population until pressurization
N_{film}	- bubble population for film boiling pressure
P_{surf}	- pressure at surface of drop
P_{trans}	- pressure sensed at transducer
P_i	- pressure of i th component v - vapor, ℓ - liquid, g - gas in metal
$\dot{q}/A$	- heat flux
R	- radius to outer oxide layer
r	- radius of vapor bubble
r_{drop}	- radius of metal drop assumed spherical
T_{fi}	- initial fuel temperature
T_{ci}	- initial coolant temperature
T_I	- interface temperature
T_{crit}	- critical temperature of coolant
T_H	- temperature of fuel at some time
T_ℓ	- liquid temperature (usually T_{ci})
T_{sat}	- saturation temperature
T_O	- surface temperature of oxide layer
T_s	- temperature of surface of tin with oxide layer
t_g	- growth time of bubble from D_A to D_B

t_w	- waiting time before bubble growth
$\langle v \rangle$	- average drop velocity
V_v	- vapor velocity
V_l	- liquid velocity
W_e	- weber number
α	- thermal expansion coefficient
α_i	- thermal diffusivity of ith material
δ	- thickness of oxide layer
σ_f	- fracture stress of tin oxide
σ	- surface tension
ρ_i	- density of ith material
τ_D	- dwell time
λ	- wavelength of disturbance at liquid vapor interface
λ_c	- collision frequency

1. INTRODUCTION

In the area of fast reactor safety the hypothetical core disruptive accident (HCDA) is one of the methods used to investigate the inherent safety of the Liquid Metal Fast Breeder Reactor (LMFBR). The two low probability accidents which are considered are the transient overpower (TOP) or the loss of flow (LOF) without scram. In the TOP failed fuel rods release molten fuel to the coolant channel where a thermal fuel coolant interaction (FCI) occurs. The result is a vaporization of the sodium and its expansion as the working fluid in the accident. In the LOF the fuel vapor is the working vapor fluid because at the time of fuel failure, the core is essentially voided of sodium liquid. In either case the end result is the expansion of the vapor with the possibility of inflicting mechanical damage on the reactor vessel and possibly breaching the containment of the radioactive inventory.

The amount of disruptive work done on the vessel in the TOP depends upon the amount of thermal energy released from the fuel in an FCI that is transformed into coolant work energy. Past analysis of postulated FCI's have predicted [1] a theoretical maximum mechanical energy conversion efficiency of around 30%. This though took no account of the physical mechanism by which this large work energy conversion could be accomplished. Extensive experimentation

and analysis has continued until the present day on this subject.

In experiments using actual reactor materials [2] and reactor environment temperatures, no energetic FCI's have been observed when hot UO_2 was dropped into cold sodium. Small pressure pulses and extensive UO_2 fragmentation did result. At some universities in the United States and laboratories in England [3][4][5][6][7][8] simulant materials of tin (fuel) and water (coolant) were used to study the FCI phenomena. In these experiments fragmentation as well as energetic pressure pulses ($\Delta P > 10$ bars) were recorded from the dropping of tin into water. This raised the question whether a common mechanism of fragmentation exists for the two sets of materials or whether the mechanistic phenomena in each case are different. Henry and Fauske of Argonne National Laboratory [9, 10, 11, 11, 13] proposed an energetics model based on analysis and experiments using simulant materials of Freon and mineral oil. One of their criteria for an energetic FCI is that the interface between the hot and the cold liquids must be above the spontaneous nucleation temperature of the coolant. This condition is satisfied for the tin-water system but not the UO_2 -sodium system.

There are many other theories as to possible mechanisms for a FCI. It is the intent of this study to review the theories presented for a FCI with a special emphasis on the spontaneous nucleation theory of Henry and Fauske. The reason for this emphasis is that this theory has never been directly

applied to the tin-water simulant data to determine if the two are consistent. The experimental results of many investigators in tin-water systems will also be reviewed to give a physical insight into the observed dependencies of the interaction.

When the model is applied to the tin-water system the data appear to be consistent with the main criteria of the model. The main difficulty in applying the model is that the bulk effects in the experiment can easily mask the local interactions on which the model is based. The many mechanisms contained in a FCI are complex and it is nearly impossible to identify all the combinations of conditions which are sufficient for a large scale FCI. The criteria of Henry and Fauske provides a necessary set of conditions for an energetic FCI which are applicable in the reactor environment or in these simplified experiments.

2. EXPERIMENTAL TIN/WATER DATA

A fuel coolant interaction involves a number of possible processes: vaporization of the coolant at a large rate, fragmentation of the fuel into small particles, generation of pressure pulses due to rapid vaporization, quiescent cooling of the fuel in film boiling without fragmentation. An inherent part of the possible consequences of an FCI is the conceivable mode of contact between the fuel and coolant.

Some modes are:

a) Jetting

A mechanistic way of involving fuel and coolant is by fuel rod failure in the core and molten fuel jetting into a coolant channel filled with liquid sodium. Both in pile Treat test and out of pile tests by Argonne National Laboratory have been done in this prototypic geometry. Although the fuel-coolant mass ratio is similar to that used by the Hicks Menzies [1] calculation, low energetic FCI's have resulted.

b) Shock Tube

Experiments examining this mode of contact visualize the reentry of sodium coolant into an area of molten fuel. The fuel could have originated due to a blockage in the coolant channel and subsequent voiding and fuel rod failure. Also the shock tube geometry gives a means for insuring direct initial contact between the fuel and coolant.

c) Static

This type of contact is similar to that experienced by Long [14] when he investigated the aluminum and water explosions. There is no movement between the fuel and coolant when the interaction begins.

d) Drop Contact

This is the type of contact in which all tin/water data has been accumulated. The possibility of free falling drops of molten fuel in the hypothetical accident exists. One reason to experimentally use this mode of contact is that the mechanisms which could cause the fuel coolant interactions are not easily disturbed by hydrodynamic effects.

The subsequent discussion then will be limited to the drop mode of contact.

Experimental data for tin dropped into water over a wide range of coolant and fuel temperatures has been accumulated [3,5,6,7]. Tables 2.1 gives a listing of the thermo-physical properties of tin, water, UO_2 and sodium as a point of comparison and information. The overall effects of the tin-water experiments illustrate that the consequences of a fuel coolant interaction depend upon a few variables: initial coolant temperature, initial fuel temperature, size of fuel mass, shape of fuel mass and the type of trigger which initiates the interaction.

2.1 Experimental Dependence on Temperature

To examine the dependencies of fuel and coolant temperatures consider Figure 2.1. The area inside the triangular region (Temperature Interaction Zone TIZ) represents the combination of initial fuel and coolant temperatures where a "noticeable" FCI occurred for tin (12 gms) being dropped in water by Dull force [5]. The initiation of the interaction was self triggered. Percentage disintegration (PD) was the means Dull force used to record the violence of the interaction.

It is defined as the ratio of the minutely fragmented mass to the original mass. To get a physical feeling of this method of measurement Figure 2.2 is presented to show the type of fragmentation that is typical when a fuel coolant interaction occurs as the initial fuel temperature is varied. The experimental procedure was standardized to obtain as reproducible results as possible. Tin was heated in a helium atmosphere to minimize oxidation and dropped into the water over a 3 cm distance. The boundaries on the TIZ represent where the percentage disintegration fell to zero and no interaction occurred. Vaughan [15] described the condition of no interaction as tin falling to the bottom of the test tank and quietly solidifying.

The left vertical cutoff in Figure 2.1 ($T_f \approx 300^\circ\text{C}$) is where the tin surface begins to solidify upon coming into contact with the water. The melting temperature (T_m) of tin is 271°C . The bulk temperature of the tin can be higher than T_m because with liquid-liquid contact the interface temperature immediately decreases to the value

$$T_I = \frac{T_{fi}}{1+\alpha'} + \frac{\alpha' T_{ci}}{1-\alpha'} \quad (2.1)$$

$$\alpha' = \sqrt{\frac{K\rho C_c}{K\rho C_f}} \quad (2.2)$$

$$\alpha' \approx .205 \text{ for Tin-Water.}$$

So when $T_I = 271^\circ\text{C}$, T_{fi} corresponds approximately to the 300°C cutoff temperature observed.

The diagonal cutoff is much more difficult to explain. A possible explanation [5][6] has been that the tin when entering the water experiences film boiling with such a thick stable film that it falls to the bottom of the test tank without interaction and peacefully solidifies. This explanation seems to be born out by the Stevens and Witte [16] experiments (using a moving solid silver sphere in water) which also exhibited a thick stable film boiling mode at these elevated levels of fuel and coolant temperatures. A final note to Figure 2.1 is that it represents a necessary but not a sufficient set of conditions for a FCI within the temperature interaction zone. So it is possible to have fuel and coolant temperatures within the TIZ and not have a self-triggered interaction.

Figure 2.3 again illustrates the TIZ but now with three other sources of experimental data included. Board [6] investigated the relationship of externally triggered interactions to the body of data in the self triggered mode. What he demonstrated was that at a fuel and coolant temperature where no self-triggered interaction had occurred, an applied external pressure pulse provided an interaction. This was done by dropping a tin drop into water and letting it come to rest on the bottom of the test tank and then applying the pressure pulse. A similar test where the ambient pressure of the water tank was suddenly increased produced the same results. In both cases Board concluded that the pressure pulse hastened the destabilization of film boiling and

provided liquid contact for the FCI to occur. Bjorkquist [7] Shiralkar [17] and Bjornard [8] at MIT performed similar experiments to those at Culham but with smaller tin masses (.8 and 6.6 gm). Measurement of the violence of the interaction was by pressure transducer measurements about 4-1/4 inches below the surface of the water bath. The tin was heated in three different types of atmospheres (air at 1 atm, nitrogen, vacuum 10^{-5} in H_2O and flushed with nitrogen). No noticeable pressure pulse differences were observed for fixed T_{fi} and T_{ci} with the three different heating environments. The variance of the peak pressure pulse during the interaction was measured as a function of the initial fuel temperature with coolant temperature held constant. The results as shown on Fig. 2.3 indicated a peak pressure pulse maximum was reached at $T_{fi} = 700^\circ C$ for a fixed coolant temperature ($23^\circ C$). A similar peak pressure pulse was observed for the experiments of Arakieri [3] at UCLA. Figure 2.3 indicates that for two fixed fuel temperatures ($T_{fi} = 787^\circ C, 676^\circ C$) a maximum peak pressure pulse was detected at a coolant temperature of $23^\circ C$ and $40^\circ C$. It should be mentioned that in all UCLA experiments the tin was heated in air and the mass of the tin was 25 gm.

All the MIT tin dropping tests which produced a pressure detected interaction had a similar qualitative pressure profile as Fig. 2.4 illustrates. Initially when the tin was dropped into water nothing occurred. A dwell time accompanied by a

a small high frequency cycle was observed. When the interaction did occur the frequency of recorded pressure peaks slowed to a few hertz and the peak pressure denoted by the small box on Fig. 2.4 was obtained. This peak pressure is plotted in the subsequent graphs to typify the violence of the interaction. Finally as the fragmentation of the hot tin becomes complete the pressure frequency increases and decreases in magnitude.

The effect of variance of the fuel temperatures on the peak pressure and percent disintegration is shown in more detail on Figs. 2.5 and 2.6. For a fixed coolant temperature of 23°C the peak pressure of the interaction goes through a maximum at 700°C. It should be noted that this is the pressure recorded at the transducer which was located about 4-1/4 inches below the water surface. The distance from it to the tin droplet varied due to the different dwell times. So the actual pressure at the surface of the drops is probably greater. Bjornard [8] indicated that it could be estimated by the equation;

$$P_{\text{surf}} = P_{\text{trans}} \left[\frac{D_D - \tau_D \langle V \rangle}{r_{\text{drop}}} \right] \quad (2.3)$$

where r_{drop} - radius of spherical drop of mutual mass
 D_D - depth of the pressure transducer
 τ_D - dwell time before interaction
 $\langle V \rangle$ - average falling velocity

This would tend to shift the recorded points to slightly higher values. As will be shown later the dwell time

increases with increasing fuel temperature. So the peak pressures from tin drops at lower initial fuel temperatures could be slightly higher at the drop surface than hotter drops. If this functional dependence is correct, the actual pressure of the tin drop increases slightly. Shiralkar [17] attempted to verify the linear dependence of this expression by experiments. No conclusive results were determined.

The percentage disintegration (PD) has a different behavior with the variance of fuel temperature. Fig. 2.6 shows for a fixed coolant temperature of 65°C that PD increases to a maximum near 100% ($T_f \approx 550^\circ\text{C}$) and remains at that level until it reaches the diagonal cutoff of the TIZ. This is a much different behavior than the peak pressure exhibited. Also note that not all the tin droppings within the TIZ produced an interaction by observed fragmentation. Again the combination of fuel and coolant temperatures within the TIZ is a necessary but not a sufficient condition for an interaction.

The data compiled by Arakeri [3] at UCLA illustrates the variance of peak pressure with the initial coolant temperature. These are pressure measurements at the transducer. The depth of the transducer from the surface of the water was estimated to be about 5-1/4 inches from the UCLA drawings. This would have a tendency to increase the actual surface pressures slightly in relation to the MIT data. As the figure indicates the same peak pressure behavior occurs

and a maximum is reached at $T_{ci} = 40^{\circ}\text{C}$ ($T_{fi} = 676^{\circ}\text{C}$) and $T_{ci} = 23^{\circ}\text{C}$ ($T_{fi} = 787^{\circ}\text{C}$). The large black dot is the representative maximum point shown in Fig. 2.3.

The behavior of the percentage disintegration with a variance in coolant temperature is illustrated in Fig. 2.8. Again the tendency to saturate near 100% is seen when $T_{ci} \approx 50^{\circ}\text{C}$. There is also the distinct cutoff at the diagonal of the TIZ.

2.2 Effect of Mass Size and Shape on Interaction

The effect of the variance of mass on possible fuel coolant interactions has been observed in other industries. Nelson [18][19] noted that in smelt (NaCO_3)-water interactions a minimum mass existed below which no interaction occurred. Additionally he maintained that the shape of the hot fuel in the drop contact mode also affected the chance of an interaction. When a continuous stream of fuel ($T_{fi} \approx 1300^{\circ}\text{C}$) was poured into water ($T_c \approx 20^{\circ}\text{C}$) thermal explosions were more numerous and of greater violence than when an initially spherical drop was used. The experiments carried out were mainly qualitative so no pressure data were obtained. This dependency on shape will be further investigated in a subsequent section.

The observation of a minimum mass for no interaction was also noted by Reynolds [4]. The interesting point was that a minimum mass of 4 gms was reported for no noticeable FCI whereas MIT [7] used a tin mass of .8 gm and obtained an interaction. This discrepancy has not been resolved.

The effect that the amount of mass has on the peak pressure derived from an interaction is shown in Fig. 2.9. As the mass decreases in quantity the peak pressure observed from the interaction increases. This observation of the data trends can be coupled with the independent observation of many investigators [3,5,6,7,20] that the number of peak pressure cycles decreases with a decrease in the mass. This second observation is explained by the fact that with less mass the amount of fragmentation of the fuel and energy release to the coolant is less. Thus fewer peak pressure cycles will occur to result in the same final fragmentation.

2.3 Dwell Time and Destabilization of Film Boiling

The dwell time is the waiting period between the time the tin drop enters the water and the time the interaction begins. For the MIT and UCLA experiments the interaction initiation was recorded by film and pressure transducer measurements. For the Culham laboratory tests a movie film was used to determine when the interaction began. The results are presented in Figs. 2.10 and 2.11 for variable initial fuel and coolant temperatures. As the coolant or fuel temperature increase the dwell time before interaction also increases. A physical explanation of this behavior could be advanced. As the tin is dropped in the water it entered into a film boiling mode of heat transfer with the water. As the temperature of the coolant or fuel is increased this mode of film boiling becomes more stable and the thickness of the film increases. This demands a longer time for the drop to cool to the point

where the interaction occurs, thus the dwell time increases.

Some related film boiling studies were conducted by Stevens and Witte [16] and may have some relevance to the tin-water system. The experiment passed a hot solid silver sphere through water at a constant velocity (1.5 m/s). The temperature of the silver could be detected by a thermocouple as it was being cooled by the water and a movie film of the transient cooling indicated the nature of the film boiling. From their experiments a heat transfer rate versus temperature of the silver could be calculated (Figs. 2.14 and 2.15) as well as a heat transfer coefficient. As a first estimate of the temperature of the tin surface at the time of the interaction, Bjorquist [7] used this heat transfer data. The results are shown on Figs. 2.10 and 2.11. Over the range of temperatures considered (400-1000°C) the heat transfer coefficient does not vary significantly ($.5-1 \text{ watt/cm}^2\text{°C}$). To obtain this estimate the tin was assumed to be in the form of a sphere. This analysis indicates that except for fuel temperatures below 400°C the average temperature at the tin surface at the time of the interaction is in the range of 500-600°C.

Additionally Stevens and Witte noted three distinct types of film boiling depending upon the initial combination of fuel and coolant temperatures. Fig. 2.12 illustrates the regions of film boiling noted by Stevens and Witte overlayed on the TIZ. The first type of film boiling recognized was a smooth thin film boiling. The important point noted was that at the time of film collapse this smooth film appeared to collapse coherently

("transplosion") and enter into pulsational and nucleate boiling. Whereas for higher initial fuel and coolant temperatures the film took on an irregular shape. Also the method of film destabilization was noticeably different. Instead of a complete coherent collapse the film collapsed incoherently in selected spots on the silver surface, until nucleate boiling resulted. Finally at very high temperature combinations a thick film developed which did not destabilize during the time the sphere was moved through the water in their experiment.

The interesting correlation is that the maximum peak pressure detected by MIT and UCLA for the tin-water system corresponds to the point where the mode of film destabilization was identified to go from a coherent to an incoherent mode. This suggests that the mode of film destabilization has a distinct effect upon the pressure derived from an interaction. The incoherency of the film destabilization process could contribute to the dissipation of the potential destructive work energy of the interaction. The attempt to correlate the results of these two experiments must be tempered by the fact one is a liquid-liquid system while the other a solid-liquid system. It is not completely clear what are the significant differences or similarities in these two systems. The behavior of film boiling with the liquid-liquid system will be discussed further in later sections but this initial comparison is worth noting.

In the next section of this investigation an overview

of the theoretical models for an FCI are examined. Special attention is given to the spontaneous nucleation theory of Henry and Fauske, because it represents an attempt not to describe the whole character of a FCI but just the necessary criteria for an energetic one. To illustrate the applicability of the theory Henry has done fuel-coolant experiments using the simulant materials of Freon and mineral oil. Fig. 2.13 illustrates that even with these materials a characteristic pressure maximum is attained at a particular fuel and coolant temperature. The results of their theoretical curve shown will be discussed later. The intent here has been to show the similarity in the experimental results and emphasize the different dependencies of the interaction on experimental parameters.

Table 2.1

Some Material Properties

	Tin[7] (liquid) (270-700°C)	UO ₂ [] (solid) (<2800°C)	Water [] (liquid) (20°C)	Sodium[][] (liquid) (1000°C)
Density (g/cc)	6.8	11	1	.896
Specific Heat ($\frac{\text{W-sec}}{\text{gm } ^\circ\text{C}}$)	.24	.46	4.18	1.25
Thermal Conductivity ($\frac{\text{W}}{\text{cm } ^\circ\text{C}}$)	.325	.021	.00643	.77
Melting Temp.	271°C	2800°C	--	--
Homogeneous Nucleation Temp.	--	--	305°C	1900°C
Critical Temp.			375°C	2130°C

Figure 2.1. Temperature Interaction Zone for 12 gm of Tin

Figure 2.2. Sequence of Pictures of Debris as the TIZ is crossed with MIT-UCLA data.

Figure 2.3. Temperature Interaction Zone data.

Figure 2.4. Typical Pressure Trace for $\text{Ti}-\text{H}_2\text{O}$ Interaction.

Figure 2.5. Peak Pressure as a Function of Initial Fuel Temperature.

Figure 2.6. Fragmentation as a Function of Initial Fuel Temperature.

Figure 2.10. Dwell Time and Interaction Temperature as a Function of Initial Coolant Temperature.

Figure 2.9. Peak Pressure as a Function of Initial Tin Mass.

Figure 2.11. Dwell Time and Interaction Temperature as a Function of Initial Fuel Temperature.

Figure 2.12. Comparison of Stevens-Witte Data to TIZ.

Figure 2.13. Peak Pressure as a Function of Fuel Temperature for Freon-Oil System.

Figure 2.14. Heat Transfer Coefficient for a Sphere in Water Inferred from Stevens-Witte [17].

Sphere Material: Silver
Sphere Velocity: 1.52 m/sec
Pool Temperature: 24 °C

Figure 2.15. Heat Flux from a Hot Sphere in Water [7].

3. PROPOSED ANALYTICAL MODELS FOR FUEL COOLANT INTERACTIONS

The mechanisms proposed for the description of fuel coolant interactions (FCI) are rather numerous. It is the intent of this section to give an overview of the different models suggested and dwell at length on the spontaneous nucleation model by Henry and Fauske [9][10][11]. It is this model which is employed in the following section to investigate the experimental findings previously presented.

To do this, the proposed models may be classified into five areas: hydrodynamics in which the fragmentation observed is due to hydrodynamic effects between the fuel and coolant independent of the thermal conditions of the two constituents; solid shell models which consider the thermal cooling effects and propose fragmentation due to these induced stresses; molten droplet models which see fragmentation as a result of pressure forces created within the coolant by bubble growth or other processes; violent gas release model where the mechanism for fragmentation is viewed as an abrupt and disruptive release of dissolved gases from the fuel into the coolant.

A propagation model has been suggested by Board and Hall [40][41]. This model assumes an energetic fuel coolant interaction has been initiated and focuses analysis on the propagation through the fuel-coolant medium. At the present

time there is considerable discussion as to the validity of some of the assumptions and analysis of the model by Bankoff and Cho [42][43][45]. There is no relevance of this model to the tin-water data to be analyzed so it will not be discussed further in this study.

3.1 Hydrodynamics Models

There are two general mechanisms considered in this area. If the fuel has large inertial forces when it comes into contact with the coolant [i.e., $W_c = (\rho D < V >^2 / \sigma)_{\text{fuel}}$ is high] then the initial impact could cause fragmentation regardless of thermal conditions. Cronenberg [23] in his review of this model notes that the independence of thermal conditions has not been substantiated experimentally. Cho [24] noted that thermal effects had a large effect on the degree of fragmentation even when the Weber number of the fuel was kept low (< 20). Thus the thermal condition can dominate the interaction. The influence of hydrodynamic effects are more notable in the jetting mode of contact, but even in this situation Cho asserts that the thermal effects are an equally important consideration.

The second mechanism considered is that the movement of the molten fuel through the coolant and high relative velocities causes wavelike disturbances to appear on the surface and break up the fuel as well as encapsulate the coolant in it for subsequent vaporization and fuel fragmentation. The initiation of fuel fragmentation has occurred at relatively small velocities [7] which seems to indicate

that this mechanism as a prime initiating mechanism is not operative.

3.2 Violent Gas Release Model

Epstein [25] has proposed a mechanism for fragmentation based upon the release of dissolved gases from the fuel. The model assumes that a liquid contains a concentration (C_g) of dissolved gases at equilibrium which follow the relation

$$C_g(P,T) = P_g^{1/2} A \exp(-\Delta H/RT) \quad (3.1)$$

where A is a constant, P_g is partial gas pressure, ΔH is the enthalpy change across the dissolution reaction of the gas and T is the system temperature. This behavior is illustrated in Figure 3.1. The model proposes that the fuel at high molten temperatures contains dissolved gas at a certain pressure (point A3). When it is cooled down rapidly, the change in temperature causes a large latent pressure driving force (A3 to B3) within the fuel because now the concentration of dissolved gas is supersaturated. The subsequent release of this gas is then seen as the mechanism for fragmentation. It should be noted that the concentration of gas increases with increasing temperature and pressure. Also note that for the same change in temperature (A1 to B1 or A' to B') the latent driving pressure is much higher when the initial pressure of the system is higher. The application of this model to the UO_2 -Na system is difficult because solubility characteristics of Xenon, Krypton and Helium dissolved in UO_2 are not well known. It is known though

that oxygen exhibits this type of dissolution behavior.

Shiralkar [17] examined this model using the tin-water system as simulant materials. Oxygen was considered as the likely dissolved gas to cause fragmentation. Tin was heated in a vacuum (10^{-5} in H_2O) to reduce the dissolved gas content to a minimum and then dropped in water under a nitrogen atmosphere. As previously mentioned this procedure reduced the latent driving pressure orders of magnitude and according to the model should have resulted in much lower fragmentation and observed pressure pulses when interacting with water. As Fig. 3.2 illustrates when results of these tin experiments are compared to similar experiments for tin heated in air, the pressure pulses are not smaller but of the same magnitude. Also the magnitude of the pressure pulses decreases noticeably at elevated fuel temperatures ($> 700^\circ C$). Reference back to Fig. 3.1 also shows that as the temperature of the fuel increases the concentration of dissolved gas goes up thus increasing the latent driving pressure. This would indicate that the model predicts more fragmentation and larger pressure pulses as the fuel temperature increases. But as Fig. 3.2 shows the observed pressure reached a maximum and falls rapidly as T_{fi} increases past $700^\circ C$. So for both of these reasons Shiralkar concluded this mechanism was not dominantly operative in the tin-water system.

3.3 Solid Shell Model

A model for a FCI suggested by Cronenberg [26][31] is that UO_2 upon contact with sodium starts to solidify and the associated thermal gradient in the solidifying shell induces stresses in the solid which can cause fragmentation. Knapp [27] extended the model by considering UO_2 as a brittle material and showed that UO_2 could fragment in the reactor temperature environment based upon a brittle failure criterion. This brittle failure analysis was utilized by Corradini [29] to illustrate that the model predicts minimum particle sizes of fragmentation near those experimentally reported [30]. In addition Lenz [28] illustrated that the rate of fragmentation is limited by the rate of solidification of the molten drop. This model would not apply to the simulant materials of tin and water because tin has a low melting point (271°C) and in the experiments performed was probably molten at the time of the interaction with water. More importantly tin is a ductile material and it would probably not be subject to brittle fracture due to thermal stresses, because plastic deformation would mitigate the consequences.

3.4 Molten Drop Models

When tin is used as the simulant fuel, interactions occur in temperature ranges where the tin is molten. Also as the tin drop enters the water coolant film boiling is the initial mode of heat transfer between the two materials. In contrast to the benign interactions of UO_2 dropped into sodium, this system undergoes an interaction which not only

produces fragmentation but large pressure pulses. This might suggest that the mechanism which is operative in these energetic interactions is not a concern in the UO_2 -sodium system. The models presented below could be thought of as explaining the mechanism that causes this inherent difference in these two different material combinations.

3.4.1 Internal Acoustic Cavitation Model

Watson et al. [34] has suggested that the behavior of the tin droplet during film boiling could be the key to its subsequent fragmentation. As it enters the water film boiling is the mode of heat transfer that occurs. Kazimi [33] has shown that a spherical drop in film boiling experiences pressure oscillations in the film due to the unstable growth and collapse of the film around the droplet. Watson hypothesized that these external pressure oscillations in the film could be transmitted into the interior of the drop and cause large internal negative pressures, resulting in the fragmentation of the tin that has been experimentally observed.

Although this theory appeared plausible, Watson [34] noted that an overall energy balance of this process results in a final fragmented particle size about an order of magnitude too large in comparison to what has been experimentally observed. Additionally the problem was originally conceived considered a perfectly spherical drop with an assumed film thickness (10^{-4} - 10^{-5} in). These simplifications allowed the oscillation of the film without any asymmetry effects

contributing to the result. These simplifications as Watson [34] concludes must be relaxed before it is finally concluded that the model can indeed be considered as a possible fragmentation mechanism.

3.4.2 Vapor Bubble Growth - Cyclic Model

The physical picture that is associated with this model is a cyclic process of coolant bubble growth and collapse on the molten fuel surface causing the observed fragmentation. Vaughan [37] describes the proposed view of the fragmentation process in three stages.

Trigger--1st Stage--Liquid-Liquid Contact

When the tin enters the water film boiling keeps the two materials from liquid-liquid contact. After a short dwell time the interaction occurs. Board [6][38] maintains that the interaction occurs when the film boiling mode of heat transfer has destabilized and liquid-liquid contact is restored between the constituents. The trigger which allows this intimate contact can vary. Recall that Board [6] experiments showed an increase in the ambient pressure or a pressure pulse initiated the interaction of a tin droplet sitting in water. His explanation is that these pressure increases collapsed the film and promoted the contact. Self-triggering of the interaction is shown experimentally by Culham Laboratory [5] and MIT [7]. Reynolds [4] interprets this self-trigger mode as the point where the tin cools down to such a temperature that the heat flux is too low to sustain the film and it subsequently collapses. It has been

estimated by the use of Stevens-Witte data that the tin surface at the time of interaction is within a temperature range (500-600°C). So the initial stage of film collapse is readily agreed upon by many experimenters.

Trigger--2nd Stage--X-Mechanism

Once the contact between liquid fuel and coolant has been made some X-mechanism causes the first bubble to grow in the coolant near the fuel surface. This initial growth continues until the bubble has expanded into a subcooled region where the pressure within the bubble decreases below ambient and the collapse begins. This X-mechanism is not specifically identified. In fact Dullforce and Buchanan [35][36] in their analysis just assume the initial bubble's presence and investigate the pressure generation in the cyclic stage. Vaughan [37] has suggested some possible mechanisms:

- i) The possibility of dissolved gas within the drop has been shown by Shiralkar [17] not to be the dominant mechanism for the fragmentation. But in this context it is possible that diffusion of dissolved gas from the fuel could provide a preferred nucleation site for the initial bubble growth.
- ii) Due to possible viscous instabilities between the coolant and the fuel, a chunk of fuel may be torn away from the large mass. Coolant could then rush in between these two fuel masses and nucleate a vapor bubble at a high rate from some vapor which has been trapped from the destabilized film.
- iii) Instead of a bubble nucleating due to the initial presence of a vapor pocket, nucleation could occur spontaneously at an elevated temperature above T_{sat} . Fauske's theory [9][10] on this will be presented next, but it suffices

to say here that the bubble nucleates at a high superheat temperature due to local density fluctuations in the coolant.

- iv) The very process of film collapse could be the mechanism for bubble growth. As the film collapses on the molten droplet, the surface could be distorted and the fuel fragmented to some extent by Taylor instabilities. This situation with some trapped vapor from film collapse could promote rapid bubble nucleation.

This stage of the model is left vague and the dynamics of the cyclic bubble growth and collapse are investigated in detail. In contrast in the spontaneous nucleation theory, the initiating events are the prime features described. This difference allows the spontaneous nucleation theory to be based on a few criteria, in contrast to the mathematical detail of this dynamic model.

Cyclic Bubble Growth and Collapse

As the initial bubble starts to collapse it is still attached to the molten fuel surface. This causes a certain asymmetry to the collapse motion and the result is a jet of coolant being accelerated toward the fuel surface. This penetrates the fuel surface and rapid spreading of the coolant within the fuel results. This interpenetration allows the nucleation of another bubble which grows out again and the sequence continues this cycle until the fuel has been fragmented and cooled to such a point that it cannot continue. Buchanan's [35] analysis suggests that the pressure increases with each cycle by the ratio

$$\frac{P_i - P_o}{P_{i-1} - P_o} = \begin{matrix} 6.673 \text{ heterogeneous nucleation} \\ 2.899 \text{ homogeneous nucleation} \end{matrix} \quad (3.2)$$

where P_0 is the system pressure.

The features of this model are that a small initial disturbance can propagate into a large interaction. The model attempts to explain the experimentally observed upper diagonal cutoff of the Temperature Interaction Zone (Fig. 2.1) by suggesting that a condition of thick stable film boiling exists whereby the tin has time to cool and solidify before the film destabilizes and the jet cannot penetrate the solid shell to escalate the interaction. The UO_2 -sodium system would not experience such an interaction primarily because film boiling is precluded. As soon as UO_2 encounters sodium, solidification begins and nucleate boiling can occur at the solid surface. As it has been previously stated it is proposed that fragmentation in this system is due to induced thermal stresses.

There are though some experimentally observed facts that the cyclic model fails to describe. As Fig. 2.4 shows the peak pressure occurs after a dwell time but at the start of the cyclic interaction, and then trails off. In the cyclic model the pressure builds up after every cycle which is exactly the opposite behavior to what is observed experimentally. Also no attempt has been made to analyze the effect of shape or mass of the drop on the violence of the interaction. The major characteristic of the model is a detailed description of the dynamics of the cyclic process, and Dullforce [36] notes the pressure response of the analysis is strongly dependent on parametric constants in the model.

Therefore the model gives a good detailed view of the cyclic interaction yet does not propose an initial set of initiating criteria which allows one to judge if a fuel coolant interaction can be precluded.

3.4.3 Spontaneous Nucleation Model

A different approach to the phenomenon of fuel coolant interactions has been made by the spontaneous nucleation theory. In contrast the model of Henry and Fauske [11] establishes the initial criteria for a FCI by a mechanistic analysis of the process of film boiling in a liquid-liquid system. The net result is a model which presents a simple but unique view of the phenomenon of film boiling. To understand the model, the concept of homogeneous and spontaneous nucleation temperatures will be reviewed briefly.

When a liquid is heated to its saturation temperature corresponding to the ambient pressure, a small amount of liquid superheat is needed to initiate boiling in the liquid at preferred nucleation sites such as solid surface cavities or pockets of dissolved gas at the solid-liquid interface. In the total absence of such preferred sites such as a clean liquid-liquid system, the liquid can still be heated to higher superheat temperatures in this metastable state. If the temperature is raised to a high enough value thermal fluctuations occur in the liquid and there is a finite probability that a cluster of molecules with vapor-like energies could come together to form a vapor embryo of the size of the equilibrium nucleus

$$r = \frac{2\sigma}{P_V - P_\ell} \quad (3.3)$$

The process of vapor nuclei formation in a metastable liquid due to density fluctuation is called homogenous nucleation, and the temperature at which the probability of such activity becomes significantly large in the homogenous nucleation temperature. The probability of the formation of these vapor nuclei can be estimated from Boltzman's equation for the distribution of molecular nuclei of size r

$$N(r) = N \exp \left[- \frac{\Delta G(r)}{kT_\ell} \right] \quad (3.4)$$

where N is a constant approximately equal to the number density of the liquid, k is Boltzman's constant and T_ℓ is the temperature of the liquid being heated. The free energy of formation $\Delta G(r)$ of a vapor bubble of size r is given by

$$\Delta G(r) = 4\pi r^2 \sigma - \frac{4}{3} \pi r^3 (P_V - P_\ell) \quad (3.5)$$

Now if equation 3.3 is substituted into the above equation for r , the following expression is obtained

$$\Delta G(r) = \frac{16}{3} \frac{\pi \sigma^3}{(P_V - P_\ell)^2} \quad (3.6)$$

To obtain the rate of vapor bubble nucleation (J) at T_ℓ due to homogeneous nucleation the collision frequency (λ) of the molecules is multiplied by equation (3.4) to obtain

$$J = \lambda N \exp \left[- \frac{16 \pi \sigma^3}{3 \Delta P^2 k T_\ell} \right] \quad (3.7)$$

The collision frequency is a function of temperature and is given by the relation

$$\lambda_c = k T / h \quad (3.8)$$

where h is Planck's constant. The value of the terms multiplying the exponential are approximately constant

($10^{32} \text{ cm}^{-3} \text{ s}^{-1}$). Where the frequency (J) becomes large ($10^3 - 10^7$), this delineates the homogeneous nucleation temperature.

The possibility of nucleation at the interface between the hot liquid and the cool one must be considered. Here in a liquid-liquid system in the absence of any other nucleation sites, the wetting between the hot and the cold liquid can affect the level of superheat necessary in the cold liquid before nucleation will occur. Skirpov [52] denotes the rate of nucleation at the interface as

$$J = \lambda N \exp \left[- \frac{16 \pi \sigma^3 f(\theta)}{3 (\Delta P)^2 k T_\ell} \right] \quad (3.9)$$

where $f(\theta)$ is a functional relationship involving the contact angle θ between the liquids and depends on the wetting characteristics of the system. Generally $f(\theta)$ is a function of time and temperature of the system ($0 \leq f(\theta) \leq 1$). For a well-wetted system ($\theta = 0$) equation (3.9) reduces [$f(\theta) = 1$] to the homogeneous nucleation form. Thus $f(\theta)$ can reduce

the temperature at which vapor pockets can nucleate spontaneously at sufficiently large rates ($J \approx 10^3 - 10^7$). So in general the spontaneous nucleation temperature is less than the homogeneous nucleation temperature, due to imperfect wetting [$f(\theta) < 1$] at the liquid-liquid interface and preferential nucleation there.

Henry [12] has noted that water does not have good wetting characteristics compared to other fluids such as sodium, so the spontaneous nucleation temperature (T_{SN}) will be less than the homogeneous nucleation temperature (T_{HN}). The capability to identify $f(\theta)$ is not attainable at present so the model and its subsequent use will be based on T_{HN} as the known upper limit for spontaneous nucleation.

The basis of the Henry-Fauske model is the establishment of a set of criteria which are necessary for an energetic fuel coolant interaction. Before a detailed view of the model is presented it would be beneficial to summarize the four requirements:

- i) There must be an initial premixing of the fuel and coolant (Fig. 3.3). This can be accomplished by film boiling being the initial mode of heat transfer between the constituents. This would allow the fuel to non-explosively become intermingled with the coolant.
- ii) Direct liquid-liquid contact is required, implying a breakdown in the film layer between the two fluids. This destabilization is dependent upon the surface temperature of the hot fluid as well as seen later.
- iii) The interface temperature once liquid-liquid contact is achieved must be greater than the

spontaneous nucleation temperature for an explosive interaction to occur, but less than the critical temperature of the coolant. This results in fragmentation and mixing of the hot and the cold fluid without delay.

- iv) For the interaction to escalate to a large scale FCI, the proper inertial constraint is also required. This somewhat nebulous criterion can be thought of qualitatively as a condition where a non-energetic interaction is characterized by the fuel and coolant being driven apart from the initial vapor explosion whereas an energetic FCI is one where fuel and coolant are more fully intermixed [41].

The basis of the model is the determination of the interface temperature between the coolant and fuel when in contact. This is given [39] by the formula in equation (2.1). If T_I of equation (2.1) is above T_{HN} then vapor pockets can nucleate. However nucleation will not occur immediately because a thermal boundary of sufficient size must grow in order that nuclei can form and grow.

Consider a cold drop impinging upon a hot fluid surface (Fig. 3.4). Upon initial liquid-liquid contact the temperature profile within the drop for short times ($10^{-10} < t < 10^{-3}$ sec) can be given by [39]

$$\frac{T_I - T}{T_I - T_C} = \operatorname{erf} \frac{X}{2 \sqrt{\alpha_c t}} \quad (3.10)$$

This can be approximated by

$$\frac{T_I - T}{T_I - T_C} \approx \frac{X}{2 \sqrt{\alpha_c t}} \quad (3.11)$$

for times less than 10^{-4} sec. In addition at the time of

contact the heating of a small liquid layer of the coolant will cause a local density decrease. This decrease causes a local expansion and pressurization. The pressure wave will have to travel to the free surface and back before pressure is relieved. So even if the initial T_I is greater than T_{HN} a waiting time of

$$t_{w1} = 2\ell/c \quad (3.12)$$

is needed where ℓ is the characteristic distance to the free surface ($\ell = \epsilon D_D$; $0 < \epsilon < 1$). During this time the thermal boundary layer continues to grow according to equation (3.10). After pressure relief the possibility of vapor nucleation exists. This can only happen if the local temperature is sufficiently high to support a bubble of the critical size [equation (3.3)] as Fig. 3.5 depicts. These sample curves are for Freon (coolant) in oil (fuel), as given by Henry [11]. Notice that at 10^{-7} sec once the vapor nuclei is formed it can grow stably to point B without collapse.

The frequency of vapor nucleation is the product of the frequency of nucleation per unit volume and the volume from the interface to the point of interest per cm^2

$$\text{Freq} = \int_V J \cdot dv \quad (3.13)$$

The waiting time then for a vapor nuclei to form once the pressure is relieved is

$$t_{w2} = \frac{1}{\int_V J dv} \quad (3.14)$$

So this is the total waiting time to a vapor bubble nucleation at the smallest critical size. When the bubble does nucleate a second local pressurization occurs due to the volume change caused by its nucleation. This pressure must be relieved by a second wave, giving a waiting time also equal in magnitude to equation (3.12). So the total waiting time before nuclei can grow is

$$t_{wT} = \frac{4\ell}{c} + \frac{1}{J \cdot V} \quad (3.15)$$

where $V = 1 \text{ cm}^2 \cdot D_A$ considering slab geometry. Note that $\int_V J dV$ has been approximated by JV . In this instance the magnitude of $1/JV$ is so small that it is insignificant in comparison to the pressure relief times. Once the bubble starts to grow, the time of growth from D_A to D_B can be approximated by the inertial bubble growth equation.

$$t_g = \frac{D_B - D_A}{2\sqrt{\frac{2}{3} \frac{P_B - P_A}{f_\ell}}} \quad (3.16)$$

This description gives a simplified view of the birth and growth of a homogeneously nucleated bubble. The physical question to ask is can the cold and hot liquid stay in intimate liquid-liquid contact. In other words, given this mechanistic view, what is the condition for film boiling versus the wetting mode of contact for the coolant on the fuel.

When the bubble has started to grow from point A to

point B, other bubbles are nucleating simultaneously. The maximum number of bubbles which nucleate during the growth of the first one is just

$$N_{\max} = J \cdot V \cdot t_g \quad (3.17)$$

This maximum population of vapor nuclei may never be reached because the rapid nucleation of bubbles during time t_g may pressurize the cool liquid locally and suppress any further nucleation. To determine this bubble population (N_{limit}) consider the change in density due to the change in volume of a constant volume liquid drop.

$$\Delta \rho_l = - \frac{\Delta V_l}{V} \rho_l \quad (3.18)$$

The change in density is related to the sonic velocity by

$$C^2 = \Delta P / \Delta \rho \quad (3.19)$$

Now the change in volume of liquid is caused by the nucleation of the bubbles, given by

$$-\Delta V_l = \Delta V_v = N_{\text{limit}} \pi D_A^3 / 6 = N_{\text{limit}} D_A^3 / 2 \quad (3.20)$$

by this method N_{limit} can be related to the relative pressurization to suppress further nucleation. Substituting equations (3.20) and (3.19) into (3.18) the expression obtained is

$$N_{\text{limit}} = 2 \Delta P / \sigma_l D_A^2 C^2 \quad (3.21)$$

This is an approximate analysis for a complex phenomenon.

So as an order of magnitude estimate Henry [11] maintains that the ΔP overpressure can be from 2 to 15 bars and not vary the conclusion drastically (e.g., a typical range for $N_{\text{limit}} = 10^9 - 10^{10}$).

The possible population of vapor bubbles has been specified as either N_{max} or N_{limit} , whichever is the smallest. To decide whether or not this population will grow and coalesce into a film or just grow isolated from one another until they collapse and rewet the surface; consider Fig. 3.6. Henry suggests that film boiling will occur if the initially nucleated bubbles grow to size D_B which occupies the entire surface. So the number of bubbles per cm^2 is

$$N_{\text{film}} = \frac{1}{D_B^2} \quad (3.22)$$

So a drop of cold liquid will enter into film boiling if N_{limit} (or N_{max}) N_{film} and will continue to wet the surface if N_{limit} (or N_{max}) N_{film} .

The finally physical point to consider is what happens if the interface temperature reaches or surpasses the critical temperature (T_{critical}) of the coolant. First the amount of molecules needed to make up a vapor bubble decreases rapidly as T_{crit} is approached (Table 3.1). Secondly there cannot be any nucleation after the critical point is past because the vapor and liquid are one in the same. Thus it appears that the critical point of the coolant is the upper temperature limit above which a FCI cannot occur. Once the

coolant contacts the hot fuel, film boiling is the immediate result since there cannot be liquid-liquid contact. Based on this logic, Henry [11] estimates that the smallest bubble that can be nucleated as T_{critical} is approached is given by

$$\frac{T_I - T_{\text{critical}}}{T_I - T_c} = \frac{D_b'}{\sqrt{4\alpha_c t_{w1}}} \quad (3.23)$$

where D_b' is an arbitrary small bubble diameter assumed by Henry to contain 40 molecules, and t_{w1} is the acoustic relief time for the initial pressure wave.

Now given this criteria for film boiling it is apparent that whether the coolant will enter into film boiling rather than wet the fuel surface is dependent on the temperature of the constituents and the initial diameter of cold drop. Henry varied parametrically the drop diameter for the Freon-oil system and obtained the plot seen in Fig. 3.7. This demonstrates that as drops get bigger film boiling will be the mode of contact and in agreement with equation (3.23) film boiling is the only mode of contact above the coolant critical temperature. This simple mechanistic analysis of the requirements of film boiling presents a mode of stable film boiling (Fig. 3.8) different than the traditional view, in which the film separated the cold and hot surfaces completely and the mode of heat transfer was due to evaporation of coolant into the film. The picture that this analysis suggests (Fig. 3.9) is that the hot surface is always being

tested by the liquid coolant in the form of coolant drops attempting to wet the surface. Only when the combination of temperature and drop size are correct will the surface be wetted by the drops. This picture is simplified by this discussion of a liquid-liquid system because no preferred nucleation sites exist to complicate the analysis. The model explains the possibility of energetic fuel coolant interactions by maintaining that they are only possible when $T_{HN} < T_I < T_{crit}$ and when the coolant drop can wet the fuel surface. If the drop is too big it will go into film boiling and evaporate or fragment itself to the size for wetting the surface. After it has wet the surface, a large amount of heat can be transferred to the coolant until the homogeneous nucleation point is reached for a large portion of the drop at which time a violent nucleation process will occur and fragment the cold drop and fuel surface. The time of heat transfer (t_{HT}) is governed by approximately the relief time of the initial pressurization due to contact

$$t_{HT} = \frac{2\ell}{c}$$

Henry approximates the amount of heat transferred by the semi-infinite slab formula [39] of

$$\frac{Q}{A} = 2 k_H (T_H - T_I) \sqrt{\frac{t_{HT}}{\pi \alpha_H}} \quad (3.24)$$

Once the process of homogeneous nucleation occurs, the drop vaporizes and a pressure pulse due to this bubble nucleation

occurs. This pressure is predicted by taking the saturation pressure of the coolant drop corresponding to the equilibrium temperature that it has been heated to by the fuel.

$$\frac{Q}{A} = \rho_c C_{p_c} \ell (T_{\text{sat}} - T_c) \quad (3.25)$$

Thus the relation is obtained

$$T_{\text{sat}} = T_c + \frac{2k_H (T_H - T_I)}{\rho_c C_{p_c} \ell} \sqrt{\frac{t_{HT}}{\pi \alpha_H}} \quad (3.26)$$

Throughout this analysis ℓ has been a parameter linked to the original drop diameter ($\ell = \epsilon D_D$, $0 < \epsilon < 1$). Henry for the analysis based on Freon-oil experiments estimates ℓ by assuming the drop doubles its projected area when it wets the fuel surface ($\ell = \frac{2}{3} D_D$). This model was compared to experiments and Fig. 2.13 shows the results which Henry concluded exhibit reasonable agreement.

This model will be utilized in the following section to evaluate the tin-water experiments. The essence of the model then is to delineate a set of criteria by which an energetic FCI is possible. The minimum temperature for film boiling in a liquid-liquid system is proposed to be T_{SN} . Additionally an energetic FCI can only occur in the narrow window between T_{SN} and T_{critical} for the coolant. So the whole concept of film boiling, destabilization and vapor explosion are linked to the spontaneous nucleation temperature.

Table 3.1

Homogeneous Nucleation Frequency as a Function of
Temperature for Water

$$P_l = 1 \text{ atm}$$

Temp. (°C)	P_v (psi)	σ (10^{-3} lb _f /ft)	D (Å)	J (cm ⁻³ s ⁻¹)
100	14.7	4.032	23,000	0
150	67	3.3	3,900	0
290	1087	1.155	91	10^{-24}
302	1274	.987	67	$4.3(10^{-5})$
305*	1324	.9	58	$3.8(10^6)$
310	1429	.827	49	$6.3(10^{15})$
315	1541	.75	42	$7.5(10^{21})$
320	1659	.674	35	$8.7(10^{25})$
330	1849	.526	25	$1.13(10^{30})$
340	2130	.389	15.5	10^{32}
355	2529	.202	6	10^{32}
370	3090	.03	.8	10^{32}

*This is approximately the homogeneous nucleation temperature for water at one atmosphere.

Table 3.2

Homogeneous Nucleation Frequency as a Function of
Temperature for Freon-22 [11]

Temp. (°C)	P_v (psi)	σ (10^{-4} lb _f /ft)	D (Å)	J (cm ⁻³ s ⁻¹)
50	281	3.2	98	$1.8(10^{-12})$
52	294	3.05	88	$5.5(10^{-3})$
54	307	2.89	80	$1.3(10^6)$
56	323	2.71	72	$7.96(10^{10})$
58	336	2.55	64	$1.75(10^{16})$
60	352	2.39	58	$4.2(10^{21})$
65	391	1.99	44	$6.2(10^{27})$
70	435	1.61	32	$6.8(10^{30})$
75	481	1.23	22	$1.5(10^{32})$
80	522	.89	14	$5(10^{32})$
85	585	.561	8	$9.2(10^{32})$

Table 3.3

Homogeneous Nucleation Frequency as a Function of
Temperature for Sodium

Temp. (°C)	P _v (psi)	σ (10 ⁻³ lb _f /ft)	D (Å)	$\frac{J}{s^{-1}}$ (cm ⁻³ s ⁻¹)
1830	1616	1.62	84	2.3(10 ⁵)
1835	1638	1.59	82	6(10 ⁷)
1840	1653	1.55	80	9(10 ⁹)
1860	1754	1.41	68	1.5(10 ⁷)
1880	1841	1.28	59	1.7(10 ²²)
1900	1943	1.14	49	1(10 ²⁵)
2000	2483	.458	14	5(10 ³⁰)

Figure 3.1. Dissolved Gas Concentration as a Function of Gas Pressure.

Figure 3.2. Peak Pressure as a Function of Initial Fuel Temperature in a Vacuum.

PREMIXED ZONE OF FUEL IN
COOLANT

Figure 3.3. Premixed Zone of Fuel in Coolant.

Figure 3.4. Pictorial View of Cold Drop Impinging on Hot Fuel Surface.

Figure 3.5. Temperature as a Function of Distance from Interface [11].

FILM BOILING MODE

$$N_{\max} \text{ or } N_{\text{limit}} > N_{\text{film}}$$

WETTING AND CAPTURE

$$N_{\max} \text{ or } N_{\text{limit}} < N_{\text{film}}$$

Figure 3.6. Pictorial View of Film Boiling and Wetting and Capture.

Figure 3.7. Criterion of Wetting of Fuel (Oil) Surface by Coolant [11].

Figure 3.8. Typical Heat Flux vs. Temperature Curve.

Figure 3.9. Two Different Views of Stable Film Boiling.

4. APPLICATION OF SPONTANEOUS NUCLEATION THEORY TO THE TIN-WATER EXPERIMENTS

The experimental range of tests in the tin-water system is quite extensive as Fig. 2.1 demonstrates. To apply the spontaneous nucleation theory to the data requires investigation into each of the specified criteria of the theory. The first criteria of initial premixing of fuel and coolant before an interaction occurs is satisfied in these experiments because the tin enters the water in a film boiling configuration which separates the two liquid constituents during the dwell time. The comparison of the other criteria to the experimental results is not as straightforward and will be dealt with individually.

4.1 Trigger: Direct Liquid-Liquid Contact

The view of the model in regard to film boiling is very mechanistic. In a liquid-liquid system the lower limit of stable film boiling is the spontaneous nucleation temperature. Above this temperature film boiling is possible depending on the size of the liquid droplet attempting to wet the surface of the fuel-coolant interface. As Fig. 3.6 depicts as the size increases the chance of film boiling increases. So in contrast to the traditional view of no contact in the stable film boiling regime, the cold liquid is always testing the hot fuel surface by characteristic droplets trying to wet it. Additionally the model suggests that film boiling is the only mode of contact above the coolant's critical temperature.

In the tin-water system all the energetic interactions occurred when the temperature of the fuel apparently greatly exceeded the critical temperature of water. As a rough cut analysis in section 2 the temperature at the time of interaction was estimated to be between 500-600°C (Figs. 2.10-2.11). At that time it was considered that the film had destabilized allowing liquid-liquid contact and the onset of the interaction. This at first glance appears to contradict the criteria of the model, but the underlying assumptions of this short analysis must be examined. The tin droplet was assumed to be spherical and chemically pure. In other words the effect of the shape of the mass and possible tin-oxide formation was ignored in this estimate. In section 2 it was noted that shape was observed to have an effect in smelt-water explosions so consideration of this variable is not unreasonable. These variables will now be examined to see if they significantly effect the possible conclusions the data can give. The objective of this investigation is to determine if the molten tin actually does cool down to surface temperatures which correspond to the temperature window $(T_{SN} < T_{surf} < T_{critical})$ where the coolant can wet the surface and engage in an energetic interaction.

4.1.1 Effect of Shape on Destabilization Temperature

In the tin-water experiments done at UCLA [3] a stroboscopic photographic method was used to observe the detail movements of the tin before and during the interaction

(Fig. 4.1). The tin enters the water bath in a distorted shape approximately spherical. As it falls through the water initially in the film boiling mode, a projection starts to grow out of the bulk of the tin. This behavior was noted time and again in these photographs. As it progressed out (usually near the bottom half of the bulk of the tin), the interaction started at this projection after the characteristic dwell time. A dark cloud probably signifying tin fragmentation engulfed the projection as the interaction propagated up towards the bulk of the tin fuel. The start of the interaction at the projection suggests that film boiling is destabilized initially in this region and proceeded up through the bulk of the fuel.

Remember that the spontaneous nucleation theory's criteria is that film boiling can only be destabilized below the critical temperature of the coolant. A possible explanation of the experimental observations is that the tin projection from the bulk of the fuel acts as a cooling fin. This fin then can cool down quicker than the bulk during the dwell time to a temperature below $T_{critical}$ and allow film destabilization. Two simple ways to approximate the growing projection are illustrated in Figs. 4.2 and 4.3. Appendix B gives the details of the analysis. The variable quantity in the analysis is the shape of the fin in relation to the bulk of the fuel. From the UCLA photographs it was assumed that the ratio of the fin diameter to a spherical drop diameter was .2. This approach was applied to the

MIT [7] tin-water data. The reason for the use of this data is that both peak pressure pulses and dwell times were measured for all the tin dropping experiments. As Tables 4.1 and 4.2 illustrate the predicted surface temperatures of the projection at the time of the observed interaction is slightly above the critical temperature of water. Remembering that the coolant when it attempts to wet the surface lowers the interface temperature further to the value of

$$T_I = \frac{T_H}{1+\alpha'} + \frac{\alpha' T_C}{1+\alpha'} \quad (2.1)$$

the surface temperature does fall below the critical temperature of the coolant at the time of the interaction.

The next question to ask is what causes the projection to appear in the tin drop experiments. One possible explanation is that the viscous forces would tend to distort or fragment the drop. As Appendix C shows the Weber numbers for all the tin experiments are not much larger (16-100) than We_{crit} and it might be possible that the drop is distorted into this shape but not fragmented by these forces. The second possibility is that an oxide layer forms on the tin surface due to initial air heating or movement through the steam space in film boiling. Since SnO_2 has a high melting point ($\sim 1800^\circ C$) it's very likely that a solid oxide layer would form on the outer surface of the tin. Then due to thermal gradient and the associated contraction, stresses could build up in this thin outer skin and crack it, dis-

charging the internal bulk of the tin in the form of a projection. This second possibility is discussed in the next section with a complete analysis given in Appendix D.

4.1.2 Effect of Oxidation on Tin-Water Experiments

There is very little information available on the rate of oxidation in tin. Dickens [47] conducted some experiments on a number of metals at temperatures ranging from room temperature to 800°C. The results of tin oxidation at room temperature over a long period of time (until no more oxide build-up was detected) were very small-- $\sim 3\mu\text{g}/\text{cm}^2$. As the temperature was increased to give a molten tin state, the amount oxidized increased linearly. The method used to measure oxidation was that tin was suspended on a microbalance as a oxygen rich gas was blown across it. The oxide which was formed was reported to be of non-coherent nature. This seems to imply that more oxide would form on some areas of the surface than others and it would easily fragment and become like floating slag on the molten tin surface.

Henry [46] has suggested that an oxide layer as well as the growth of a projection could explain how the tin surface cooled down to such an extent that the interface temperature would be below T_{critical} for the water. Appendix D explains a simple analysis that indicates that the thickness of oxide ($\sim 2\text{mm}$) needed to reduce the surface temperature close to T_{critical} for water is just not realistic in comparison to the experimental maximum oxide thicknesses observed.

The possibility of thermal stress fragmentation as a means of causing the observed projection growth was also examined. Appendix D covers the analyses in detail while the model and the results will be discussed here. The physical picture being considered is a molten sphere of tin surrounded by a very thin shell of tin oxide (SnO_2). The temperature gradient across this thin shell is assumed to be linear. As Appendix D illustrates the temperature across the shell is less than a 1°C , but because the shell is so thin its fracture is likely. The constant properties stress equations as given by Kent [48] are used to determine the tangential stress component which can cause cracking. The criterion used to judge if the computed stress will cause fracture is based on the fracture stress of tin oxide ($\sim 17,000$ psi) as reported by Lynch [49]. The computed tensile stress at the surface of the shell was about 10^5 psi. This greatly exceeds the fracture stress, and seems to confirm the fact that the tin oxide will crack. This analysis seems to indicate that the growth of the observed projection on the tin drop could be caused by an outer shell of tin oxide cracking. As Knapp [27] illustrated the cracking is due to the thermal and pressure stresses induced by the thermal gradient across the shell. The induced stresses could push a portion of the molten tin outward in the form of a projection.

The overall comparison of the data to the spontaneous nucleation model seems to indicate the likelihood that the

tin is cooled to a surface temperature such that

$$T_{SN} < T_I < T_{crit}$$

when film destabilization occurs. This would lend support to the second criterion of the model in establishing an upper temperature bound on the point of film destabilization.

The important concept to remember in the model is that film destabilization is literally the wetting of the hot fuel surface by small water droplets. At the same time it is these droplets which are the source of the observed pressure pulse from the ensuing interaction. This is the restatement of the third criterion of the model and will be applied next to the tin-water data.

4.2 The Interaction as Depicted by the Spontaneous Nucleation Theory

When the tin surface has reached a temperature near the coolant critical temperature destabilization can occur. The process that makes this possible are random contacts by small water droplets on the tin surface. This physical picture is suggested by the model although there are other possible interpretations. Bankoff [44] has suggested that the contact could actually be in the form of tongues of liquid projecting from the bulk of the liquid to test the hot surface. Again these tongues have a randomized size which corresponds to the characteristic depth λ used by Henry. Using the same procedure as outlined in section 3.4.3 a plot can be constructed of the regions of film boiling and

surface wetting and capture as a function of characteristic diameter D ($\ell = D$ assumed) and interface temperature (Fig. 4.5). This plot is useful in outlining the region in which drop diameters can wet the tin surface to cause the interaction. Appendix E gives a further description of how the graph was constructed.

A difficulty now arises in the application of the model to the tin-water data. Unlike the small local Freon-oil experiments of Henry [12] when the cold drop diameter could be controlled and photographically observed, these experiments were bulk tests where effects tended to mask the details of the interaction. No such observation could occur here. Instead all that was measured in these experiments as section 2 describes was the pressure history and the degree of final fragmentation. Many other variables had a possible effect in these experiments. Shape and oxide formation effects have been suggested but most likely the full famifications of all the variables cannot be addressed. In addition the most random and unpredictable event is the intermittent contact during film boiling. Because of these interwoven effects it is doubtful the model is useful in predicting the observed pressure as equation (3.26) was meant to do. The random wetting of the surface by these drops occurs on a microscopic scale and it is the coherence of these many interactions that gives the observed pressure behavior. To use the macroscopic data to predict this pressure by the model does not seem warranted.

What can be done is the reverse analysis. The only amenable experimental data to use is the peak pressure pulse during the interaction. This can be used to evaluate the characteristic depth λ which is needed to produce the pressure for the interaction. The peak pressure pulse is used in this analysis because it occurs immediately at the beginning of the interaction period after the dwell time. Physically this could be envisioned as the time when the initial population of cold water droplets of sufficiently small size wet the tin surface. After a characteristic time, spontaneous nucleation occurs and generates a pressure pulse. This is probably the most coherent phase of the interaction because the tin/water configuration is quite distinct, and thus produces the greatest pressure. As mentioned in section 2 another inherent uncertainty is the difference between the detected transducer pressure and that at the drop surface. Bjornard [8] suggested a relation between them but Shiralkar could not experimentally confirm it. After this initial interaction the fuel and coolant in the vicinity is fragmented further and most likely more water droplets are created which again wet the tin surface and continue the cyclic process, until the thermal fuel energy is expended.

Henry suggests that the pressure possible from such an interaction corresponds to the cold liquid saturation pressure after the fuel has transferred its heat to the coolant. The simple energy balance on a drop of volume V is given as

$$\rho_l C_{p_l} V (T_{c_f} - T_{c_i}) = 2k_H A (T_H - T_I) \sqrt{\frac{t_I}{\pi \alpha_H}} \quad (4.1)$$

Henry simplifies this to the form

$$\Delta T_c = \frac{2k_H (T_H - T_I)}{\rho_l C_{p_l} l} \sqrt{\frac{t_I}{\pi \alpha_H}} \quad (4.2)$$

where $V/A = l$ the characteristic depth and t_I is proportional to the relief time ($4l/c$). When these relations are combined the result is

$$\Delta T_c = \frac{4k_H (T_H - T_I)}{\rho_l C_{p_l} \sqrt{\pi \alpha_H l}} \quad (4.3)$$

Given the experimentally measured pressures for the MIT and UCLA tests, the characteristic depth can be determined and are depicted in Table 4.3-5 and Figs. 4.6-4.8. The details of the analysis are given in Appendix E. The results indicate two points. The size of characteristic water drops ($l \equiv D$) all reside in the wet and capture region boundary of Fig. 4.5; thus this simple analysis is self-consistent. Secondly, to match the shape of the peak measured pressures the characteristic depth decreases to a minimum at the maximum pressure. To try to understand why this probably occurs in this model consider equation (4.1) again,

$$\rho_l C_{p_l} V (T_{c_f} - T_{c_i}) = 2k_H A (T_H - T_I) \sqrt{\frac{t_I}{\pi \alpha_H}} \quad (4.1)$$

The two major assumptions made to proceed to equation (4.3) are that the time of heat transfer and the V/A ratio

are both proportional to the drop size ($D_D \approx \ell$). This seems like a reasonable approximation for the time t_I because it must be associated with D so that the pressurization wave generated by the droplet contact is relieved ($t_I = 4\ell/c$) and vapor generation occurs in a short enough time span to propagate the interaction. But the ratio V/A may not necessarily follow the diameter's (D) variance. Physically $\rho_\ell C_{p_\ell} \frac{V}{A} \Delta T_c$ is the amount of energy deposited into the coolant which manifests itself by vaporization in the form of a pressure source per unit surface area of contact. It is possible that the volume of coolant contributing to the pressure source is not proportional to characteristic depth (ℓ) but depends upon the initial conditions of the film boiling mode. That is the ratio V/A could depend on the coherency of the film boiling mode and the coolant droplet wetting.

Stevens and Witte [16] noted (Section 2.3) that the macroscopic mechanism for film destabilization which they observed could be classified by either transplosion or progressive instability, and the degree of each type's behavior was a function of the initial fuel and coolant temperatures. This idea could be applied here and pointed out that V/A might depend on the coherency of the hydrodynamics at the time of destabilization and that this is highly temperature dependent. If the behavior described by Stevens and Witte is applied here then the coherency of film destabilization noticeably decreases as the temperature surpasses 700°C.

Stevens and Witte denoted this coherent/incoherent transition from transplosion to progressive instabilities at the same point where the maximum pressures for tin/water were detected and a significant peak pressure decrease subsequently occurred. This behavior could be the reason for the variance of ℓ with initial fuel temperature. If the characteristic depth results are replotted qualitatively (Fig. 4.9) with a separation of ℓ and $V/A = d$ then this distinction might be more easily seen. Although the characteristic depth of the droplets (ℓ) may not change with temperature (T_{f_i}) the relative coherency of their wetting the tin surface and collectively contributing to the sensed peak pressure pulse may change with T_{f_i} . This conclusion seems to be consistent with the observations of Stevens and Witte and may help in understanding the peculiar tin-water pressure behavior.

Finally, this concept of coherency of the interaction seems to correspond well with the final criterion of the spontaneous nucleation theory. This states that the proper physical constraint is needed for an energetic interaction. It has been observed that the largest peak pressures occurred at one fuel-coolant temperature combination. Additionally other experimentation for solid-liquid systems suggests that at this same temperature combination an observed change in the mechanism of film destabilization occurred from a coherent to a more incoherent mode. All this suggests that the final criterion on proper constraints is a function of the fuel-coolant initial temperatures as well as

the possible spatial constraints imposed. At different initial temperatures an FCI may be inherently less energetic even though it is within the temperature window of $T_{SN} < T < T_{crit}$. Fig. 2.13 showing a pressure maximum in Freon-oil experiments could be further indication of this behavior in other simulant material systems.

4.3 Experimental Characteristics not Predicted by Spontaneous Nucleation Theory

There are some characteristics of the tin-water data that do not directly seem to be explained by the spontaneous nucleation theory. The observed diagonal cutoff of the Temperature Interaction Zone does not seem to match the model. Regardless of the initial fuel temperature, given a sufficient dwell time for the fuel to cool down to the range $T_{SN} < T_I < T_{crit}$, the model suggests that the fuel should have an interaction with the coolant. This might indicate that the notion of droplets continually testing the fuel surface may be realistic but that in its simplicity it is ignoring the possible incoherencies of contact that could mitigate the extent of the interaction at high temperatures. The thickness of the film boiling vapor could increase to such an extent that very infrequent contact is made by these random water droplets, thus causing an extremely incoherent destabilization and a benign interaction. It also may be possible that the characteristic size of these droplets depends on the initial temperatures of the constituents, so that as T_{f_i} or T_{c_i} increases, the size of the droplet

increases. This would favor the film boiling mode of contact at much lower interface temperatures. Because $T_{SN} < T_{HN}$ for water, film boiling may not destabilize until the interface is below the melting point of tin. Once solidification does occur the interaction would be benign.

A second experimental effect which is not directly relatable to the model is the trend of the peak pressures to increase with a decrease in the initial mass and interaction cutoff at low tin masses. This behavior again seems to exhibit the size effects of the interaction. A possible resolution of the observed trend to the model is that as the mass increases the process of fuel surface wetting and film destabilization becomes more localized. As it becomes more localized the pressures developed by the spontaneous nucleation interaction are not globally manifested as with smaller masses, thus the observed pressure pulses decrease. This localization could be due to the increased distortions of the fuel mass as the Weber number increases.

The spontaneous nucleation theory's application to the tin-water system indicates that the criteria it sets as necessary for an energetic interaction appear to be met by the tin-water experiments. Additionally the bulk effects of the experiments tend to add more variables into the possible interaction intensity. These variables tend to contribute to the incoherencies of the interaction and mitigate its ultimate effects.

Table 4.1

Tin Cooling Analysis

MIT Data $m = 6.6 \text{ gm}$
 $D = 1.2 \text{ gm}$

T_{f_i} (°C)	T_{c_i} (°C)	h watt/cm ² °C	τ_{dwell} (msec)	Surface Temperature at Time of Interaction				
				spherical drop	spherical projection*		fin projection**	
				before water contact	before water contact	after contact	before water contact	after contact
500	23	1	5	520	438	367	445	364
600	23	.9	15	600	430	360	455	374
700	23	.75	35	610	410	344	460	377
800	23	.7	70	620	410	344	460	377
900	23	.6	100	625	375	315	455	374
1000	23	.5	150	650	370	311	460	377

* $d/D = .2$

** $d/D = .2$ $L/D = .6$

Table 4.2

Tin Cooling Analysis
Minimum Values for Cooling Projections

MIT Data $m = 6.6 \text{ gm}$
 $D = 1.2 \text{ cm}$

T_{f_i} (°C)	T_{c_i} (°C)	h watt/cm ² °C	τ_{dwell} (msec)	surface temperature at time of interaction				
				spherical projection			fin projection*	
				$K = d/D$	before water contact	after contact	before water contact	after contact
500	23	1	5	.32	450	373	445	364
600	23	.9	15	.22	448	372	455	374
700	23	.75	35	.25	452	374	460	377
800	23	.7	70	.26	448	372	460	377
900	23	.6	100	.27	453	374	455	374
1000	23	.5	150	.29	451	374	460	377

* $d/D = .2$ $L/D = .6$

Table 4.3

Calculation of Characteristic Depth (ℓ) for
Observed Pressure Pulse

MIT Data: $m_o = 6.6 \text{ gm}$

$T_{ci} = 23^\circ\text{C}$

Interface* Temp. (°C)	Initial Fuel Temp. (°C)	Average Peak Diff. Press (psi)	Corresponding Temperature Difference (°C)	ℓ (Å)
375	400	12	18	3600
375	500	22	28	1500
375	600	28	33	1065
375	700	110	73	212
375	800	35	38	800
375	900	2	4	52000
375	1000	<<1	--	--
340	400	12	18	3000
340	500	22	28	1200
340	600	28	33	860
340	700	110	73	170
340	800	35	38	630
340	900	2	4	58000
340	1000	<<1	--	--

*The difference between the two chosen is small so only 340°C is subsequently used.

2

Table 4.4

Calculation of Characteristic Depth (l) for
Observed Pressure Pulse

UCLA Data: $m_o = 25 \text{ fm}$
 $T_{ci} = 30^\circ\text{C}$

Initial Fuel Temp. ($^\circ\text{C}$)	Peak Differential Press. (psi)	Corresponding Temperature Difference ($^\circ\text{C}$)	l ($\text{\AA}$)
400	<.1	--	--
525	3	6	25000
700	22	28	1200
780	20	26	1370

Table 4.5

Calculation of Characteristic Depth (ℓ) for
Observed Pressure Pulse

MIT Data: $m_o = .8 \text{ gm}$ $T_{ci} = 23^\circ\text{C}$

Initial Fuel Temp. ($^\circ\text{C}$)	Average Peak Differential Press. (psi)	Corresponding Temperature Difference ($^\circ\text{C}$)	ℓ ($\text{\AA}$)
400	2	4	57000
500	10	16	3600
600	25	31	1000
700	180	93	110
800	60	53	330
900	10	16	3600
1000	<1	--	--

Figure 19. Selected Frames from Figure 18 Are Shown for Additional Details.

Figure 4.1. View of Growth of Tin Projection from Bulk of Tin Mass.

PROBABLE SHAPE OF
TIN DROP

APPROXIMATE MODEL WITH
SPHERE PROJECTION FOR
TIN FUEL

Figure 4.2. Model of Growing Projection Using a Sphere Projection.

PROBABLE SHAPE OF
TIN DROP

APPROXIMATE MODEL
WITH FIN PROJECTION
FOR TIN FUEL

Figure 4.3. Model of Growing Projection Using a Fixed Tin Projection.

Figure 4.4. Model of Formation of Tin-Oxide Shell on Molten Tin.

Figure 4.5. Criterion of Wetting of Fuel (Tin) Surface by Coolant.

Figure 4.6. Characteristic Depth (λ) as a Function of Initial Fuel Temperature for MIT Data.

Figure 4.7. Characteristic Depth (λ) as a Function of Initial Fuel Temperature for UCLA Data.

Figure 4.8. Characteristic Depth (λ) as a Function of Initial Fuel Temperature for MIT Data.

Figure 4.9. Qualitative Picture of Characteristic Depth as a Function of Initial Fuel Temperature.

5. CONCLUSION AND RECOMMENDATIONS

The concept of spontaneous nucleation has been shown to be applicable to the tin-water experimental data. In conjunction with this conclusion, an implication to the actual UO_2 -sodium reactor system can be made. The UO_2 sodium system appears to be a more benign combination of materials in comparison to the possible energetic interactions of the tin-water system. The reasons for this relate directly to the physically observed phenomenon in the tin-water system and the spontaneous nucleation model. Film boiling is not probable in the UO_2 -sodium because the interface temperature for this material combination and reactor environment temperatures ($900^\circ\text{C} < T_I < 1200^\circ\text{C}$) is much lower than the homogeneous nucleation temperature ($T_{\text{HN}} \approx 1900^\circ\text{C}$) for sodium. This means that the initial requirement of coarse premixing is not fulfilled and the chance of an energetic interaction is significantly reduced. In fact the most probable mechanism for the observed UO_2 fragmentation when dropped into sodium is thermal stress fragmentation as stated in Section 3 [27][28].

Thus all four criteria of the spontaneous nucleation theory have been investigated in light of the tin-water data and found to be consistent. With the consideration of the fuel shape and oxidation potential the model requirement for film destabilization within a temperature range

$T_{SN} < T_I < T_{crit}$ was found to be consistent with the data. Experimental pressure measurements were used to demonstrate that the model's characteristic drop sizes were consistent with those needed for wetting and capture and illustrated a variance in the coherency of the interaction with temperature.

The final conclusion is that usage of tin and water as simulant material in this present drop mode configuration does not appear to be warranted for future work. The bulk effects of dropping the tin into the water seems to complicate the analysis and give results with a large amount of experimental scatter. As the analysis of section 4 indicated, only a few key sets of data (pressure, dwell time) were useful in comparing the experiment to the spontaneous nucleation model. All the other observed effects could only be qualitatively linked to the model to provide any coherent understanding.

This is not to say that work should not be continued with the tin-H₂O system. One criterion in the spontaneous nucleation model is that wetting of the fuel surface is not possible when $T_I < T_{crit}$. Although the tin-water data appear to be consistent with this criterion it would be beneficial to continue experiments which look specifically at this criterion. In this instance usage of tin as the fuel might be beneficial but in a different configuration. Because of its low melting point and low vapor pressure it provides a hot fuel source over a wide range of temperatures.

A coolant (such as Freon or water) could be dropped onto the fuel in a configuration similar to Henry. From this configuration it might also be possible to obtain a physical feeling for the variance of the degree of liquid-liquid contact with temperature and possible incoherencies in the process and still have size effects included.

As past analysis has shown, fuel coolant interactions are possible outside these temperature regions. However, the resulting pressure pulses are small and the consequences of such interactions have been considered in LMFBR safety analysis.

APPENDIX A

Film Collapse Based on
Notion of Helmholtz Instabilities

A traditional way of depicting the destabilization of film boiling is to view it as the approach to a hydrodynamic instability (such as Helmholtz instability). Figure A.1 gives a pictorial view of the concept. The liquid and vapor are traveling parallel to their interface as water is being evaporated into the film space during stable film boiling. Due to the density difference the relative velocity of the vapor is much higher than the liquid. The point of film collapse could be thought of as the condition where heat flux supplies just enough energy to evaporate liquid.

$$\frac{\dot{q}}{A} = \frac{m_v h_{fg}}{A_{\text{sphere}}} \quad (\text{A.1})$$

where A_{sphere} is surface area of assumed spherical drop. As the heat flux decreases the mass flow rate of vapor decreases. Thus the relative velocity of vapor to liquid decreases. This condition can result in a Helmholtz instability. Physically this occurs when the relative velocity between the vapor and liquid becomes small enough that the vapor cannot keep the liquid from collapsing onto the hot surface. The reason that the liquid attempts to collapse is that the relative acceleration is from the less dense vapor to the more dense liquid. Schliting [50] gives the condition of instability as

$$(V_v - V_l)^2 = \frac{(\rho_l + \rho_v)^2}{\rho_l \rho_v} \frac{\sigma 2\pi}{\lambda (\rho_l + \rho_v)} \quad (\text{A.2})$$

Now in the present situation the vapor velocity is probably much larger than the liquid and liquid density is much larger than the vapor so the approximation is made

$$(V_V - V_L)^2 \approx V_L^2 \approx \frac{2\pi\sigma}{\lambda\rho_V} \quad (\text{A.3})$$

Now the wavelength (λ) at which this instability will grow is the critical wavelength of the Taylor instability given as

$$\lambda = 2\pi \sqrt{\frac{\sigma}{g(\rho_L - \rho_V)}} \approx 2\pi \frac{\sigma}{g\rho_L}^{1/2} \quad (\text{A.4})$$

Combining this with equation (a.3) the result is

$$V_V = \left[\frac{\sigma}{\rho_V} \right]^{1/2} \left[\frac{g\rho_L}{\sigma} \right]^{1/4} \quad (\text{A.5})$$

This relation can be substituted into equation (A.1) for the velocity in the mass flow rate. The result is

$$\left. \frac{\dot{q}}{A} \right|_{\text{film collapse}} = \frac{\rho_V A_{cs} h_f g}{A_{\text{sphere}}} \left[\frac{\rho_L g}{\sigma} \right]^{1/2} \left[\frac{\sigma}{\rho_V} \right]^{1/4} \quad (\text{A.6})$$

where A_{cs} is the cross sectional area of the film equal to

$$A_{cs} = \pi D \delta \quad (\text{A.7})$$

where δ is the film thickness. When the area terms are replaced the final form of the equation is

$$\left. \frac{\dot{q}}{A} \right|_{\text{film collapse}} = \frac{\rho_V \delta h_f g}{D_D} \left[\frac{\rho_L g}{\sigma} \right]^{1/2} \left[\frac{\sigma}{\rho_V} \right]^{1/4} \quad (\text{A.8})$$

To check this simple model the data of Stevens and Witte [16] are useful. They tested film boiling phenomena with a moving solid silver sphere immersed in water. They located a thermocouple in the sphere to record temperature as a function of time. By this method and movie films they were able to determine the heat flux, film thickness and temperature of the sphere at the time of film collapse. At a water pool temperature of 24°C film collapse occurred at these conditions

$$\frac{\dot{q}}{A} \approx 400 \text{ watts/cm}^2$$

$$T_{\text{surface}} \approx 450^\circ\text{C}$$

$$\delta \approx .25 \text{ mm}$$

Using the simple model derived and the experimental film thickness the predicted heat flux is (properties evaluated at 1 atm and 24°C)

$$\frac{\dot{q}}{A} \approx 20 \frac{\text{watt}}{\text{cm}^2}$$

This simple check suggests that this view of film collapse is not complete, and fails to predict this phenomena on a moving sphere. In the next few appendices the spontaneous nucleation model will provide a much better view of the phenomena.

$$\frac{\dot{q}}{A} = \frac{m_v h_{fg}}{A_{\text{sphere}}}$$

$$m_v = \rho_v A_{\text{flow}} V_v$$

$$V_v = \left[\frac{\rho_l g}{g_c \sigma} \right]^{1/4} \left[\frac{\sigma g_c}{\rho_v} \right]^{1/2}$$

Figure A.1. Pictorial View of Possible Helmholtz Instability of Film Destabilization Point.

APPENDIX B

Effect of Shape on Film Destabilization

Two approaches were used to approximate the cooling of the observed projections in the tin drop experiments.

The first was to consider the projection a sphere (Fig. 4.2) as the representation of the projection. The ratios of the sphere diameter to the overall tin diameter was assumed to be .2. This quantity was determined by visual inspection of tin drop photographs (Fig. 41.). The sphere was assumed to be initially at temperature T_{f_i} . This transient convection problem for a sphere was solved with the aid of standard plotted transient curves for various surface heat transfer coefficients. The heat transfer coefficient across the film that is employed (Fig. 2.15) in the analysis is based on results of a film boiling study done by Stevens and Witte [16]. Their tests used a solid silver sphere moving in a water bath in the film boiling mode. The water bath temperature is the same (24°C) as well as the range of hot surface temperatures (200-800°C). The time of heat transfer before interaction is based on the experimentally measured dwell time for each tin-water interaction. MIT [7] data are used for this comparison because numerous tests were made at each temperature plateau so that average values can be obtained. This hopefully is more valid than any one data point. The results are presented on Table 4.1 and illustrate that the surface temperature does indeed

cool quite significantly in the observed time. These temperatures fall within the region where the spontaneous nucleation model predicts film destabilization is possible.

The second approach to the analysis of the cooling of the projection is by considering it as a fixed fin attached to the bulk of the tin drop (Fig. 4.3). By doing this the fact that it is actually growing with time is ignored. The fin is assumed to start off with the same temperature T_{f_i} as the principal tin drop and cool down in this transient situation. The end of the fin is assumed to contribute a negligible amount of heat loss from the body ($\frac{T}{X} = 0$ at end of the fin) and temperature of the tin bulk is assumed to be essentially constant over this short span of time. Carslaw [39] gives the solution to this transient fin problem as

$$T = \frac{2(T_{f_i} - T_{c_i})}{L} \sum_n \exp(-vt - \alpha \beta_n^2 t) G_n(x) + T_{c_i} \quad (B.1)$$

where

$$G_n(x) = \cos \beta_n x \left[\frac{\alpha(-1)^n \exp(vt + \alpha \beta_n^2 t)}{(\alpha \beta_n^2 + V)} + \frac{(-1)^n}{\beta_n} \right] \quad (B.2)$$

$$\beta_n = \frac{(2n + 1)\pi}{L} \quad (B.3)$$

$$\alpha = k/\rho c \quad (B.4)$$

$$V = \frac{h P_R}{\rho c A_{csec}}$$

The ratio of the diameter of the fin to the drop diameter is again .2 and the fin length is assumed to be three times the fin diameter. Both are estimates from photographs of the UCLA tests [3]. The heat transfer coefficient is again taken from the Stevens and Witte experimental data on film boiling [16]. Since this is an infinite series solution it was found that after the seventh term for the times representative here the temperature solution did not change very much. As Table 4.2 illustrates the predicted surface temperatures by this approximation compare very well with those using the sphere projection model. It must be remembered though that when the water attempts to wet the hot surface the interface temperature is further reduced to

$$T_T = \frac{T_H}{1+\alpha'} + \frac{T_c \alpha'}{1+\alpha'} \quad (2.1)$$

$$\alpha' = \sqrt{\frac{k \rho c_{\text{coolant}}}{k \rho c_{\text{fuel}}}}$$

Considering both of these cooling mechanisms the surface temperature is below the critical temperature of water at the time of film destabilization. This is consistent with the spontaneous nucleation model.

APPENDIX C

Weber Number of Tin Water Experiments

To determine if the viscous forces are large enough to distort the tin droplet upon entering the water bath the impact Weber numbers are computed for the four different masses of tin-water experiments by $We = \rho \frac{DV^2}{\sigma}$ (C.1)

Experimenters	Mass (gm)	Diameter (cm)	Impact Velocity (cm/s)	We
MIT [7]	.8	.6	115	16
MIT [7]	6.6	1.2	115	32
CULHAM [5]	12	1.5	77	18
UCLA [3]	25	1.9	165	100

In the UCLA tests the distortion was very noticeable but no breakup occurred. The same results were reported from the other experimental sources. The possibility exists then that the initial entry into the water distorts the drop so that a fin-like projection is created.

Shiralkar [17] suggests an order of magnitude analysis that can be done to determine the time (τ) for drop deformation based upon the above parameters. The governing equation for the falling tin drop through water is

$$\rho \frac{DV}{Dt} = \frac{dp}{dr}$$

for assumed inviscid flow, where ρ = density of tin, p = pressure drop across the drop and V = velocity. In this

simple analysis the physical picture is that the pressure difference across the drop acts as the driving force to deform it. The deformation that occurs is the order of the drop radius

$$\Delta r \sim R$$

The average velocity of deformation can be approximated by

$$V = \frac{dr}{dt} \approx \frac{R}{\tau}$$

Finally the pressure is $dp = \Delta p$ across the drop. With these approximations the resultant equation is

$$\tau = R \sqrt{\rho / \Delta p} \quad (C.2)$$

This equation is a rough estimate of the time required for drop deformation through a distance comparable to R . For the experimental data by Bjorkquist [7]

$$\rho = 6.8 \text{ gm/cc}$$

$$\Delta p = 2 \text{ psi (peak pressure measured during dwell time)}$$

$$\tau = 2 \text{ msec}$$

This time is less than the dwell times for all the initial tin temperatures suggesting that there is ample time for drop deformation due to inertial forces.

APPENDIX D

Effect of Oxide Layer on Tin-Water Interaction

The likelihood of tin oxide (SnO_2) formation is very high in any system. Oxide formation can occur in the presence of air or water vapor. Dickens [47] has done some experiments on the rate of oxide formation and states the amount of build up small. In his experiments he suspended tin in an oxygen rich atmosphere on a micro balance to detect weight increase with time and blew this atmosphere over the specimen. What was found was the oxide layer formed quickly but was relatively small. The initial tin temperatures were varied from 25-800°C and a linear dependence of oxide buildup on temperature was noted. At room temperature for instance (25°C) the oxide build up was about 3 $\mu\text{g}/\text{cm}^2$. Additionally Dickens noted that the thickness of oxide build up was somewhat random with non-coherent patches observed after the experiment. This might suggest that the tin oxide which has a high melting point ($\sim 1800^\circ\text{C}$) cracked due to thermal gradient stresses on the thin shell.

Henry [46] has suggested that an oxide layer on the tin droplet may be partially responsible for causing the droplet surface temperature to fall below his theory's upper bound for film destabilization. To check this suggestion a simple analysis of a spherical tin drop at initial temperature $T_{f,i}$ and a thin layer of oxide (thickness δ) was done (Fig. D.1). The object of this short calculation was

to determine the required thickness (δ) of oxide to lower the surface temperature of the tin droplet to the vicinity of the coolant critical temperature (375°C) after the observed dwell time (τ_D). This according to the spontaneous nucleation theory would allow film destabilization to occur.

The thermo physical properties of tin oxide (SnO_2) are listed in Table D.1. The cooling analysis utilized the heat transfer coefficients (h_{cf}) for film boiling as determined by Stevens and Witte [16]. The method of calculation is as follows:

- pick an initial thickness of δ
- find the total heat transfer resistance from the molten tin surface to the coolant

$$\frac{1}{h_T} = \frac{1}{h_f} + \frac{\delta}{k_{so}} \quad (\text{D.1})$$

- with the initial temperature of tin being T_{fi} and that of the water T_{ci} find the surface temperature of the T_s molten tin surface after the experimentally observed dwell time (τ_D). A standard transient conduction chart for a sphere can be used.
- determine the heat flux out of the surface by

$$\frac{\dot{q}}{A} = h_T (T_s - T_{ci}) \quad (\text{D.2})$$

- determine the outer surface temperature of the oxide as

$$T_o = T_{ci} + \frac{\dot{q}/A}{h_c} \quad (\text{D.3})$$

- iterate by this procedure until a thickness (δ) is found which reduces the outer surface temperature to the vicinity of the coolant critical temperature.

As Table D.2 illustrates that the approximate thickness needed to accomplish this outer cooling is $\delta = .2$ cm. It should be noted again that the temperature (T_o) at the surface will be cooled further by the wetting of the water droplets on the fuel surface [according to equation (2.1)].

To see if this thickness is realistic a simple calculation can be made based on Dickens [47] experimental findings. In the range of 25°C-1000°C according to the data, the maximum oxide layer is linearly proportional to temperature ($3 \mu\text{g}/\text{cm}^2$ (exposed area @25°C) so the range of oxides layer are 36-120 $\mu\text{g}/\text{cm}^2$. The corresponding oxide thickness is found by dividing the density into these values. The result is .05 μm - .2 μm . So the actual maximum oxide thickness is four orders of magnitude smaller than what is needed to reduce the surface temperature significantly. The only conclusion is that this possibility is not realistic.

A second possible effect of the oxide layer could be that due to induced thermal stresses across this solid thin shell and pressure induced in the molten tin, the oxide layer cracks and allows tin to issue forth in the form of the observed fin projections [3]. Cronenberg [26] and Knapp [27] have investigated this possibility for UO_2 solidifying in sodium to predict fragmentation. The method of comparison is to determine the temperature profile in the solid shell and calculate the induced stress field. Knapp [27] goes on to predict based on stress intensity factors if the shell will undergo brittle failure and crack

propagation. This final analysis cannot be done with tin oxide because not enough mechanical properties are known to make a clear judgement. What can be done is to calculate the stress field and compare the maximum tensile stress at the surface with the known fracture stress ($\sigma_f = 17000$ psi) to judge if the oxide will fracture.

For simplicity the temperature field across the solid oxide shell is assumed linear. This is a good assumption here because the temperature difference is small ($\Delta T \approx .1^\circ\text{C}$). The form of the profile is then

$$T = T_s - (T_s - T_o) \left[\frac{(r - a)}{R - a} \right] \quad (\text{D.4})$$

where T_o is outer oxide temperature

T_s is temperature at oxide-tin interface.

The form of the general constant property stress equations are given by Kent [48] and Cronenberg [26] as

$$\sigma_r = \frac{2\alpha_E E}{1 - \nu} \left[\frac{r^3 - a^3}{(R^3 - a^3)r^3} \int_a^R \bar{T}(r,t) r^2 dr - \frac{1}{r^3} \int_a^r \bar{T}(r,t) r^2 dr \right] - \frac{P_a^3 (R^3 - r^3)}{r^3 (a^3 - R^3)} \quad (\text{D.5})$$

$$\sigma_t = \frac{2\alpha_E E}{1 - \nu} \left[\frac{2r^3 + a^3}{2(R^3 - a^3)r^3} \int_a^R \bar{T}(r,t) r^2 dr + \frac{1}{2r^3} \int_a^r \bar{T}(r,t) r^2 dr - \frac{1}{2} \bar{T}(r,t) \right] + \frac{P_a^3 (2r^3 + R^3)}{2r^3 (a^3 - R^3)} \quad (\text{D.6})$$

where

$$\text{temp. diff. } \bar{T}(r,t) = T_s - (T_s - T_0) \frac{r-a}{R-a} - T_m \quad (D.7)$$

$$\text{pressure stress } p = \frac{6 \alpha_E E}{R^3 + 2a^3 - \nu(4a^3 - R^3) + \frac{2E}{3k}(R^3 - a^3)} \int_a^R \bar{T}(r,t) r^2 dr \quad (D.8)$$

The tangential stress component is the one of interest because it provides the largest tensile stress at the surface of the oxide shell. To make this calculation a temperature combination which is representative of MIT tin-water data is used to illustrate the analysis. At both the beginning of the drop experiment when the surface temperature of the tin (T_s) is near 700°C or at the end when $T_s = 400^\circ\text{C}$, the temperature difference is small ($\sim .05^\circ\text{C}$). This is found by finding the heat flux

$$\frac{\dot{q}}{A} = h_c (T_s - T_{c_i}) \quad (D.9)$$

where h_c corresponds to Stevens and Witte film boiling heat transfer coefficient.

The temperature difference across the oxide shell is then

$$(T_s - T_o) = \frac{\dot{q}}{A} \frac{\delta}{k_{so}} \quad (D.10)$$

The expression for temperature [equations D.4 and D.7] can be analyti-

cally integrated in the stress equation. The oxide layer used in this illustration is .1 μm ($R - a$). The result of the calculation indicates that the tangential stress (σ_t) is approximately 10^6 psi. This is a factor of 10 larger than the fracture stress of tin oxide. So it appears that the oxide will fracture under the thermal conditions specified. Moreover the variance of T_{f_i} and the oxide thickness (δ) within the pertinent experimental ranges have no effect on the results of the analysis. Whether this fracture causes the molten tin to be pushed out in the form of a fin projection is hard to analyze. But what can be concluded is that an induced pressure is created in the tin which when the oxide shell fractures could conceivably cause the observed fin projection.

Table D.1
Properties of Tin-Oxide

<u>Property</u>	<u>Symbol</u>	<u>Reference Temperature</u>	<u>Value</u>
thermal conductivity	k_{so}	(400°C)	.22 watt/cm°k
density	ρ_{so}	(100°C)	7 g/cc
specific heat	c_p	(varies)	$17.66 + 2.4(10^{-3})T$ $- 5.16(10^5)/T$
coefficient of thermal expansion	α_E		$3(10^{-6})$ in/in°F (cal/gmole°k)
fracture stress	σ_f	(25-1000°C)	17000 psi
poisson ratio	D		.3
Youngs Modulus	E	(100°C)	$33.8(10^6)$ psi
Bulk Modulus	K	(100°C)	$28.17(10^6)$ psi
Shear Modulus	G	(100°C)	$13.(10^6)$ psi
melting temp.	T_m		1800°C

Table D.2

Tin Cooling Analysis Based on Formation of Oxide LayerMIT Data: $m_o = 6.6 \text{ gm}$ $D = 1.25 \text{ cm}$

T_{f_i}	T_{c_i}	$\frac{h_{\text{conv}}}{\text{watt}}$	$\frac{h_{\text{tot}}}{\text{watt}}$	δ	D_o	τ_{dwell}	Surface Temperature
(°C)	(°C)	($\text{cm}^2 \cdot ^\circ\text{C}$)	($\text{cm}^2 \cdot ^\circ\text{C}$)	(cm)	(cm)	(msec)	(°C)
500	23	1.0	.52	.2	1.2	5	260
600	23	.9	.5	.2	1.2	15	320
700	23	.75	.45	.2	1.2	35	360
800	23	.7	.425	.2	1.2	70	375
900	23	.6	.39	.2	1.2	100	390
1000	23	.5	.34	.2	1.2	150	450
700	23	.75	.55	.1	1.2	35	440

PROBABLE SHAPE OF
TIN DROP

APPROXIMATE MODEL
WITH OXIDE (SnO_2)
LAYER

Figure D.1. Approximate Model of Tin-Oxide Layer Formation.

APPENDIX E

Tin-Water Interaction as Predicted by
Spontaneous Nucleation Theory

To apply the spontaneous nucleation theory to the tin water system, it is necessary that the dependence of $J(T)$ on temperature is calculated. This is provided in Table 3.1. As the water droplet attempts to cool the hot fuel surface it goes through the processes described in section 3.4.3. The drop wets the surface and a density decrease due to heating causes pressurization and nucleation suppression until the wave has relieved itself. This waiting period occurs twice, the second time due to the initial homogeneous nucleation bubble. After this waiting time the bubble can grow to an equilibrium size D_B . This size depends on when the equilibrium diameter for a nucleating bubble,

$$D = \frac{4\sigma}{P_v - P_\ell} \quad (E.1)$$

intersects with the transient temperature profile of the coolant given as

$$\frac{T_I - T}{T_I - T_c} = \frac{x}{2\sqrt{\alpha_c t}} \quad (E.2)$$

This intersection changes with time. So when the waiting time (t_w) for initial bubble nucleation and growth given by

$$t = \frac{4\ell}{c} + \frac{1}{J \cdot V} \quad (E.3)$$

$$\ell = \epsilon D_D ; 0 < \epsilon < 1 \quad (E.4)$$

ends the bubble can grow to size D_B . It is at this time that the value of D_B is evaluated. In the tin water system the drop enters the water at a temperature much greater than the critical temperature of water (T_{crit}). Thus after a dwell time it is probable that an extending projection cools to a temperature near T_{crit} . After these random droplets of water have contacted the surface, the temperature falls below T_{crit} . Since this is a variable process, instead of taking the calculated interface of the tin surface which falls between T_{HN} (305°C) and T_{crit} (375°C) as calculated in Table 4.1, two representative temperatures (340°C and 375°C) are used. So a series of graphs (Figs. E.1 - E.4) are constructed to show the variance of the temperature with distance from the fuel-coolant interface. As the graphs show for the two values of T_I the value D_B does not differ drastically. In fact for the intended purposes of finding the characteristic depth ℓ it will be shown that again these two values have little difference. A second note is that the interface temperature is assumed constant during the wetting phenomenon of these droplets. This approximation is reasonable because the interaction proceeds on the time scale of 10^{-7} sec and in this time frame the interface temperature will not noticeably change.

With these curves constructed the initial drop diameter can be parametrically varied to determine at a given

interface temperature where the drop will change its final contact mode from one of film boiling to one of wetting the fuel surface. As section 3.4.3 describes this consists of determining three different bubble populations for each combination of drop diameter and interface temperature. The characteristic relief depth (ℓ) is assumed to be equal to the drop diameter for this analysis. The three different bubble populations are given as

$$N_{\max} = J D_n t_g \quad (\text{E.5})$$

$$N_{\text{limit}} = \frac{2\Delta P}{\rho_\ell C^2 D_n^2} \quad (\text{E.6})$$

$$N_{\text{film}} = \frac{1}{D_B^2} \quad (\text{E.7})$$

where

$$t_g = \frac{(D_B - D_n)}{\sqrt{\frac{2}{3} \frac{(P_B - P_n)}{\rho_\ell}}} \quad (\text{E.8})$$

Usually $N_{\max} > N_{\text{limit}}$ so that physically the population of bubbles which nucleate are limited by the fact that they overpressure the liquid coolant region and suppress further nucleation. So the criterion for the constructed Fig. 4.5 is:

- o film boiling $N_{\text{limit}} > N_{\text{film}}$ on fuel surface
- o wetting and capture $N_{\text{limit}} < N_{\text{film}}$ of fuel surface.

The final possible use of the model is to predict the peak pressure from the tin-water experiment based on the physical picture that the vapor explosion will occur only

when the drop can wet the surface.

After a pressurization relief time the process of homogeneous nucleation will cause the interaction to proceed with a pressure corresponding to the saturation pressure of the final equilibrium temperature of the coolant in that characteristic depth (ℓ). So the fuel temperature is

$$T_{C_f} = T_{C_i} + \frac{2k_H (T_H - T_I)}{\rho_\ell C_p \ell} \sqrt{\frac{t_I}{\pi \alpha_H}} \quad (E.9)$$

where $t_I \approx 4\ell/c$.

As section 4.2 pointed out the bulk effects of the tin water experiment preclude actual prediction of the pressure to compare with experiment because no information is available on the approximate size of ℓ . So the approach is taken that ℓ is found from the experimentally determined pressure pulses. Further it is suggested that the ratios V/A do not equal ℓ generally, but do equal some depth d which characterizes the coherency of the bulk interaction. So the quantity $\frac{d}{\sqrt{\ell}}$ is found and tabulated in Tables 4.3-4.5. The formula to determine $\frac{d}{\sqrt{\ell}}$ is

$$\frac{d}{\sqrt{\ell}} = \frac{4k_H (T_H - T_I)}{\rho_\ell C_p (T_{C_f} - T_{C_i}) \sqrt{\pi \alpha_H C}} \quad (E.10)$$

Figure E.1. Coolant Temperature as a Function of Distance from Interface ($t = 10^{-7} \text{ sec}$).

Figure E.2. Coolant Temperature as a Function of Distance from Interface ($t = 10^{-6}\text{ sec}$).

Figure E.3. Coolant Temperature as a Function of Distance from Interface ($6 = 10^{-5}\text{ sec}$).

Figure E.4. Coolant Temperature as a Function of Distance from Interface ($t = 10^{-4}\text{ sec}$).

REFERENCES

1. Hicks, E. P., Menzies, P. C., "Theoretical Studies on the Fast Reactor Maximum Accident," ANL 7120, 1965.
2. Armstrong, D.R., Cho, D.H., ANL 7800, "Laboratory Experiments," pp. 340-358, 1972.
3. Arakeri, V.H. et al., "An Experimental Study of the Thermal Interaction for Molten Tin Dropped into Water," UCLA-ENG-7502, December 1975.
4. Reynolds, J.A. et al., "Fuel Coolant Interactions-- Some Basic Studies at the UKAEA Culham Laboratory," CLM/RR/S2/7, SN16/2, March 1976.
5. Dullforce, T.A., et al., "Self-Triggering of Small Scale Fuel Coolant Interactions," CLM-P424, 1975.
6. Board, S.J. et al., "Fragmentation in Thermal Explosions," International Journal of Heat Mass Transfer, Vol. 17, pp. 331-338, 1974.
7. Bjorkquist, G.M., "An Experimental Investigation of the Fragmentation of Molten Metals in Water," TID-26826, 1975.
8. Bjornard, T.A., "An Experimental Investigation of Acoustic Cavitation as a Fragmentation Mechanism of Molten Tin Droplet in Water," MIT COO-2831-3TR.
9. Fauske, H.K., "On the Mechanism of Uranium Dioxide Sodium Explosive Interactions," Nuc. Sci. Eng. 51, 95-101, 1973.
10. Fauske, H.K., "Some Aspects of Liquid-Liquid Heat Transfer and Explosive Boiling," ANL, Reac. Analysis and Safety Division, 1973.
11. Henry, R.E., Fauske, H.K., "Energetics of Vapor Explosions," AIChE-ASME Heat Transfer Conf., San Frans., Aug. 11-13, 1975, 75 HT-66.
12. Henry, R.E. et al., "Vapor Explosions," with Subcooled Freon, 3rd Specialist Meeting on FCI, Tokyo, March 1976.
13. Henry, R.E., McUmbler, L.M., "Experiments on the Nucleation Behavior Under Vapor Explosive Conditions," ANS Transactions 23, pp. 370-376, 1976.
14. Long, G., "Explosions of Molten Aluminum in Water-- Cause and Prevention," Metal Progress, Vol. 71, May 1957.

15. Vaughan, G., Private Communication, October 1976.
16. Stevens, J.W., Witte, J.C., "Destabilization of Vapor Film Boiling around Spheres," Inter. Jour. Heat and Mass Transfer, Vol. 16, pp. 669-678, 1973.
17. Shiralkar, G., "An Investigation of the Fragmentation of Molten Metal Dropped into Cold Water," MIT, COO-2781-7TR.
18. Nelson, W., Kennedy, E.H., "What Causes Kraft Dissolving Tank Explosions," Paper Trade Journal, 1972.
19. Nelson, W., "A New Theory to Explain Physical Explosions," Combustion Engineering, 1972.
20. Zyszkowski, W., "Study of the Thermal Explosion Phenomenon in Molten Copper-Water System," Int. Jour. of Heat and Mass Transfer, Vol. 19, pp. 849-868, 1976.
21. Zyszkowski, W., "Thermal Interaction of Molten Copper with Water," Int. Jour. of Heat and Mass Transfer, Vol. 18, pp. 271-287, 1975.
22. Zyszkowski, W., "On the Transplosion Phenomenon and the Liedenfrost Temperature for the Molten Copper-Water Thermal Interaction," Int. Jour. of Heat and Mass Transfer, Vol. 19, pp. 625-633, 1976.
23. Cronenberg, A.W., Grohmes, M.A., "A Review of Fragmentation Models Relative to Molten UO_2 Breakup when Quenched in Sodium Coolant," ASME Conference Paper, Nov. 1974.
24. Cho, D.H., Gunther, W.H., "Fragmentation of Molten Materials Dropped into Water," Reactor Development Program Progress Report, ANL, 1973.
25. Epstein, Michael, "Thermal Fragmentation--A Gas Release Phenomenon," Nuclear Science and Engineering, Vol. 5, pp. 462, 1974.
26. Cronenberg, A.W., and Fauske, H.K., "A Thermal Stress Mechanism for the Fragmentation of Molten UO_2 in Sodium Coolant," Nuclear Eng. and Design, 1974.
27. Knapp, R., "Thermal Stress Initiated Fracture as a Fragmentation Mechanism in UO_2 -Sodium Fuel Coolant Interaction," Nuclear Eng. and Design, Vol. 35, 1975.
28. Lenz, F., "Mixing Requirements for the Limiting Fuel Coolant Interactions in Liquid Metal Fast Breeder Reactors," COO2781-8TR

29. Corradini, M., "Prediction of Minimum UO_2 Particle Size Based on Thermal Stress Initiated Fracture Model," COO-2871-4 TR, Aug. 1976.
30. Mizuta, H., "Fragmentation of Uranium Dioxide after Molten Uranium Dioxide-Sodium Interaction," Nuclear Science and Technology, VII, Feb. 1974.
31. Cronenberg, A.W., "A Comparison of Solidification Phenomena for UO_2 , UC and UN Relative to Quenching in Sodium Coolant," Nuclear Eng. and Design, 1976.
32. Watson, C.E., "Transient Heat Transfer Induced Pressure Fluctuations in the Fuel Coolant Interaction," MIT - Masters Thesis, MITNE-156, 1973.
33. Kazimi, M.S., et al., "A Theoretical Study of the Dynamic Growth of a Vapor Film Around a Hot Sphere in a Coolant," International Heat Transfer Conference, Tokyo, 1973.
34. Watson, C., Kazimi, M., et al., "Acoustic Cavitation as Mechanism of Fragmentation of Hot Molten Droplets in Cool Liquids," Future MIT - Topical Report, 1976, COO-2781-6TR.
35. Buchanan, D.J., "A Model for Fuel-Coolant Interaction," CLM-P361, Culham Laboratory, 1963.
36. Buchanan, D.J., Dullforce, T.A., "Fuel Coolant Interactions Small Scale Experiments and Theory," Second Specialist Meeting on Sodium Fuel Coolant Interaction, Nov. 23, 1973.
37. Vaughan, G.J., Caldarola, L., Todreas, N.E., "A Model for Fuel Fragmentation During Molten Fuel/Coolant Thermal Interactions," Fast Reactor Safety Conference, Chicago, Oct. 1976.
38. Board, S.J., "The Role of Spontaneous Nucleation in Electricity Generating Board, June 1974.
39. Carslaw, H.S., Jaeger, J.C., "Conduction of Heat in Solids, Clarendon Press, Oxford, 1959.
40. Board, S.J., Hall, R.W., "Propagational Thermal Explosions Part 2: A Theoretical Model," Central Electricity Generating Board, Dec. 1974.
41. Board, S.J., "Thermal Vapor Explosions," 3rd Specialist Meeting on Fuel Coolant Interaction, Tokyo, March 1976.

42. Cho, D.E. et al., "Some Aspects of Mixing in Large Mass, Energetic Fuel Coolant Interactions," Fast Reactor Safety Conference, Chicago, Oct. 1976.
43. Bankoff, S.G. et al., "Mechanisms of Vapor Explosions," Fast Reactor Safety Conference, Chicago, Oct. 1976.
44. Bankoff, S.G., Ochai, M., "Liquid-Liquid Contact in Vapor Explosions," Fast Reactor Safety Conference, Chicago, Oct. 1976.
45. Bankoff, S.G., Jo, J.H., "On the Existence of Steady State Fuel Coolant Thermal Detonation Waves," NU-2512-6, 3rd Specialist Meeting of Fuel Coolant Interaction, Tokyo, March 1976.
46. Henry, R.E., Private Communication, Oct. 1976.
47. Dickens, P. et al., "Oxidation of Metal and Alloys," Transactions of Faraday Society, Vol. 65, p. 2235, Aug. 1969.
48. Kent, C.W., "Thermal Stresses in Spheres and Cylinders Produced by Temperatures varying with Time," Transactions ASME, APM-54-18, 1932.
49. Lynch, J.F., "Engineering Properties of Ceramics," Batelle Memorial Institute, AFML-TR-66, June, 1966.
50. Schletburg, A., Boundary Layer Theory, Prentice Hall, New York.
51. Tong, L.S., Two Phase Flow, Wiley, New York.
52. Skirpov, V.P., Metastable Liquids, Halstead Press, Jerusalem, 1974.