RADIATION INDUCED NUCLEATION OF THE VAPOR PHASE

by

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(1965)

Submitted in Partial Fulfillment
of the Requirements for the
Degree of Doctor of
Philosophy
at the
Massachusetts Institute of Technology
March 1970

Signature of Author

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Chairman, Departmental Committee
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Submitted to the Department of Nuclear Engineering on March 19, 1979 in partial fulfillment of the requirements for the degree of Doctor of Philosophy.

ABSTRACT

The general theory of radiation induced nucleation is investigated for determining the threshold conditions for occurrence. A modified Volmer approach is developed in this work which treats the phenomenon from a homogeneous nucleation standpoint. This approach is more general in nature but is also more difficult to apply than the much simpler "energy balance method". It is shown that the "energy balance method" gives very good results for the conditions of interest in this work. The theory becomes completely determinate with the "cylindrical growth and instability break-up model" for determining the effective radiation track length which participates in the formation of a single critical bubble. This model appears to be applicable to all substances, types of radiation, and system conditions.

The theory is reasonably verified by experimentally obtained threshold superheats for fission fragments and fast neutrons in water at low pressure. The experimental technique involves the suspension of a drop of water in an appropriate oil to remove heterogeneous nucleation sites. Superheats of greater than 100 degrees F could be obtained and held for time periods on the order of hours.
Application of the theory to fission fragments in sodium assuming energy transferred to the electronic system of the sodium to be ineffective in bubble formation gives threshold superheats in the range of 400°F at about 100 psia. Fast neutrons would give superheats which are considerably higher. The effectiveness of this phenomenon for limiting the maximum possible superheat in a LMFBR is not encouraging.

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Acknowledgements

This thesis was started in the latter part of 1967 under Dr. Henri Fenech. In the spring of 1969, after Dr. Fenech left the Institute to assume a position at the University of California, the supervision of this thesis was undertaken by Dr. Warren M. Rohsenow. I thank both of these men for their guidance and assistance. I also appreciate the efforts of Dr. Thomas O. Ziebold who served as the reader for this thesis and Dr. Norman C. Rassmussen who's consultations were very helpful.

Much of the typing which was involved in this effort was done by my wife, Nancy. To her I express my sincerest gratitude for her understanding, encouragement, and companionship during both good and bad times. I would also like to thank Miss Clare Egan and Miss Rosemary Driscoll for their help in typing this work. I appreciate the interest of my fellow students with whom I had many helpful and stimulating discussions.

Construction of the experimental apparatus was performed in the Reactor Machine Shop. The devotion of these men to helping students and the warm friendship they provided will long be remembered. I thank all other individuals generally associated with the reactor who assisted in this effort by loaning equipment or giving advice.
The computer work required for this thesis was done at the M.I.T. Information Processing Center on the IBM 360/65.

The author was supported in his first year at M.I.T. by a National Science Foundation Traineeship. Two years of support were provided by Atomic Energy Commission Fellowships in Nuclear Science and Engineering. During the remainder of the time at M.I.T. the author held Research and Teaching Assistantships.
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Chapter I

Importance of Nucleation Phenomenon in Sodium Cooled Fast Reactors

During the past several years very considerable concern has developed over the so called "sodium void problem" in large sodium cooled fast reactors. The concern has been such that large amounts of time and effort have gone into attempts to understand and alleviate the problem. There also appears to be considerable difference of opinion over the actual severity of the problem if it, indeed, exists and what to do about it assuming it does exist. The "sodium void problem" comes about as a result of removal of the sodium from part or all of a large fast reactor core. This removal of the sodium affects the neutron balance in the core in that it effectively removes parasitic neutron absorbing material from the core, changes the neutron spectrum due to the effective removal of moderating material, and may alter the neutron leakage from the core. The first two effects tend to increase the neutron production rate and drive the reactor toward higher power and the third effect tends to drive the reactor subcritical or toward lower power. Then, depending on the relative magnitude of these effects the net result of sodium voiding may drive the reactor
supercritical or subcritical. It is, of course, the former situation that people worry about.

There are two different philosophical approaches to the problem assuming a positive "sodium void coefficient of reactivity", which is a measure of the tendency toward increasing neutron production or reactor power, does indeed exist and is sufficiently large to override any counteracting effects. The most conservative approach from a safety standpoint is to alter the design of the reactor in such a way that the void coefficient will never under any circumstances lead to a positive power coefficient. This, then, insures a "passive" power decrease in case voiding occurred. It is passive in the sense that no action by man or machine need be taken to insure against a supercritical situation. This approach does, however, degrade the performance characteristics of the reactor and result in the incurrence of an economic penalty.

The second approach is to strive for a more economic reactor and depend on detection and control systems to shutdown the reactor if voiding occurs or is imminent. This requires complete confidence in being able to detect a possible problem and, then, having complete confidence in the control system to function and function in a sufficiently short time interval.

The one most serious situation that could develop in regard to voiding comes about as a result of the ability
of liquid sodium to attain very high superheats on engineering surfaces, such as stainless steel under certain conditions. If this occurs, nucleation of the highly superheated sodium will lead to violent flashing, resulting in very sudden ejection of the sodium from the coolant channel or channels. The possibility of this occurring undetected in a few coolant channels due to some sort of flow impediment followed by possible propagation effects might lead the reactor into serious trouble under the second philosophy. The rate of sodium ejection is related to the amount of superheat as indicated by Judd (1) in figure I,1. This particular work indicates a decrease of at least a factor of two in the ejection time when the sodium flashes at superheats on the order of 200°C. Under certain conditions the reactor would respond directly to the rate of sodium ejection and could develop a serious situation in a time period inversely proportional to the rate of sodium ejection. Then, the time interval in which a control system would have to operate in order to prevent a serious problem, assuming the abnormal condition could be detected, could be considerably shortened by a highly superheated condition.

The economically attractive second approach to the sodium void problem would be considerably more attractive from a safety standpoint if the possibility of superheating the liquid sodium to a high degree did not exist at all
or if, at least, a known upper bound on the degree of superheating that could occur were known. This is what led to the investigation of radiation induced nucleation. Since a reactor is an intense source of radiation of several types, it was considered worthwhile to investigate the possibility that this radiation might place an inherent limit on the superheat obtainable in the reactor. With the question seemingly unresolved in the literature, it was decided to attempt the development of a theory for the prediction of the threshold conditions under which radiation induced nucleation would occur.
Figure I.1 Expulsion of Sodium from a Blocked Subassembly.
Chapter II

The Statistical Theory of Radiation Induced Nucleation

The statistical theory of nucleation or the theory of homogeneous nucleation is a method of describing the phenomenon of nucleation of phase changes from a microscopic point of view. Nucleation is a process in which a sufficient number of atoms of the original phase become arrayed in a configuration which is characteristic of the new phase such that the phase change can proceed without an increase in the free energy of the defined system. Homogeneous nucleation is distinguished by the fact that it takes place in the bulk of the original phase without the aid of solid surfaces, dissolved gases, or foreign particles. Even though the new phase may be more stable for a particular thermodynamic state, the process may not occur due to the existence of a potential barrier (free energy barrier). This barrier to nucleation results from the necessity of having a surface separating the two phases which requires energy for its formation. When the system is free from all outside influences this energy requirement must be fulfilled by those energetic molecules in the liquid which are capable of overcoming the potential barrier. A nucleus will be formed and nucleation will result
if a sufficient number of these energetic molecules can get together at the same location in the original phase. This nucleus is then said to be of critical size and to contain a critical number of molecules.

The effect of nuclear radiation on the nucleation of a vapor phase in a liquid is to supply part of the above energy requirement. The radiation will deposit energy in a very localized region of the liquid and cause a certain number of liquid molecules in the radiation track to take on a vapor-like configuration. This partially formed nucleus or embryo will then have a higher probability of "maturing" to the critical size than if it had to be formed completely from single molecule additions for the same system state. The problem, then, is to find the conditions under which a given type of radiation can result in the nucleation of a new phase in the original phase. More specifically it is desired to find the amount of liquid superheat which is required for a given ambient pressure and type of radiation in order to make radiation induced nucleation of a particular substance possible.

II.1 Definition of the Nucleus and the Free Energy Barrier

In order to develop this theory the concepts of free energy barrier and critical sized embryo which will
be referred to as a nucleus must be defined. The total free energy change involved in forming an embryo, which is assumed spherical, of radius $r$ is composed of two parts, a free energy change due to the formation of a surface between the phases and a free energy change associated with the transformation of a metastable state (superheated liquid) to a stable state (saturated vapor). The surface free energy change is positive since it involves placing the system in a state of higher energy. The transformation free energy change is negative since it involves the transformation of a portion of the system which is in a high energy state (superheated) to the equilibrium state.

\[ \Delta F_{\text{embryo}} = \Delta F_{\text{surface}} + \Delta F_{\text{transformation}} \quad \text{II.1} \]

\[ = 4\pi r^2 \sigma + \frac{4}{3}\pi r^3 (\Delta f_{\text{trans}}) \]

Here $\sigma$ is the surface free energy per unit surface area or the surface tension and $\Delta f_{\text{trans}}$ is the transformation free energy per unit volume. When these quantities are plotted as a function of $r$ following Brophy (2), a maximum occurs in the total free energy change at $r = r^*$ as shown qualitatively in figure II.1. The maximum free energy change, $\Delta F^*$, can be found by setting the derivative of equation II.1 with respect to radius equal to zero.
This is the free energy barrier which must be overcome if nucleation is to occur.

\[ \Delta \mathcal{G}^* = \frac{16\pi \sigma^3}{3} (\Delta f_{\text{trans}}^*)^2 \]

Figure II.1 Free Energy Considerations of Embryo Formation.

From these free energy considerations it can be seen that an embryo of radius \( r^* \) fulfills the requirement for nucleation to occur, i.e., the phase change can proceed without increasing the total free energy of the system. This embryo of radius \( r^* \) is defined as a nucleus.

The change in free energy for a critical sized embryo or nucleus can be obtained in a more explicit form.
by making use of the fact that the nucleus is actually in a state of unstable equilibrium. This requires the equality of temperature and chemical potential of the nucleus and the surrounding liquid, as shown in Hatso-
poulos (3). The introduction of the $\Omega$ potential is suggested by Uhlmann (4) which is the potential for doing work at constant temperature and chemical potential.

$$\Omega = U - TS - \sum_1 N_1 \mu_1 = - PV$$  \text{II.3}

Here $U$ is the internal energy, $T$ is the temperature, $S$ is the entropy, $\mu_1$ is the chemical potential of the $i^{th}$ component, $N_1$ is the number of moles, $P$ is the pressure and $V$ is the volume. The work required to form the critical nucleus is then the difference in this potential for a defined system of a certain mass $m$ with and without a nucleus, plus the difference in this potential for the environment in both cases. This second term is required due to the volume change in the system.

This analysis applies only to the case of a critical embryo and is actually a special case of the work of formation of an embryo of any size which is given appropriately by the change in the Helmholtz free energy of the system and environment defined in figure II.2. This free energy function is shown in equation II.4. The chemical
potential, $\mu$, for a single component substance is by definition the specific Gibbs free energy, $h - Ts$, where $h = u + Pv$.

\[ F = U - TS \]

\[ f = u - Ts \]  \hspace{1cm} \text{II.4} \]

Then, the Helmholtz function can be expressed in terms of the chemical potential as follows:

\[ f = u - Ts = \mu - Pv \]  \hspace{1cm} \text{II.5} \]

The total Helmholtz functions for the system shown in figure II.2 at states 1 and 2 are:

\[ F_{s1} = m \mu_1 - P_1 v_1 m \]  \hspace{1cm} \text{II.6} \]

\[ F_{s2} = (m - m_v) \mu_1 - P_1 v_1 (m - m_v) + m_v \mu_v - P_v v_v m_v + \sigma A_v \]

$m_v$ is the mass of vapor in the embryo and $\sigma A_v$ is the work done against the interface. The Helmholtz functions for the environment for the two system states is given by equation II.7. The change in volume of the environment is the same as the change in volume of the system which is $\Delta V = m_v (v_v - v_1)$. 
The total change in the Helmholtz free energy of the system plus environment from state 1 to state 2 gives the total work which may be done during this change of state.

\[ F_1 - F_2 = F_{s1} - F_{s2} + F_{e1} - F_{e2} \]

\[ \Delta F = -m_v(\mu_v - \mu_1) + m_v V_v(P_v - P_1) - \sigma A \]

For the special case of an embryo of critical size, \( \mu_v = \mu_1 \), and the first term in equation II.8 disappears.

\[ \Delta F^* = \frac{4}{3} \pi r^3 (P_v - P_1) - 4\pi r^2 \sigma \]

![Figure II.2 A System With and Without a Nucleus.](image-url)
The state of unstable equilibrium at \( r = r^* \) will be departed from if the embryo either grows or collapses. Then at the precise radius of \( r = r^* \) a state of dynamic equilibrium must exist. For dynamic equilibrium of the nucleus, i.e. neither growing or collapsing, a force balance on the nucleus gives a relation between the pressure difference across the interface and the radius.

\[
P^* - P_1 = \frac{2\sigma}{r^*} \tag{II.10}
\]

Putting this expression into equation II.9 gives:

\[
W^* = -\Delta Q^* = \frac{16\pi \sigma^3}{3 \left( P_v^* - P_1 \right)^2} = -\Delta F_{\text{critical embryo}} = -\Delta F^*
\tag{II.11}
\]

The free energy barrier against nucleation is now defined explicitly in terms of the state of the system. Note also that equation II.10 defines the critical radius for nucleation in terms of the state of the system. The state of the system is here established by the system pressure, \( P_1 \), and the system temperature, \( T_1 = T_v \), with \( P_v \) being the saturation pressure corresponding to the system temperature.
II.2 General Theory of Embryo Formation

In this section the general mechanism for the formation of an embryo of new phase which will be referred to as $\beta$-phase in an original phase which will be referred to as $\alpha$-phase. The system under consideration will be assumed to be at some steady state condition of temperature and pressure.

Assume, now, that this state is such that statistical variations in the distribution of the molecules give rise to a number of very small groups of molecules, i.e. a few molecules each, which could be classified as characteristic of the new phase. In the case of interest in this work these small groups of molecules would appear as vapor phase in the liquid system. The question of interest is, what is the mechanism by which these small vapor embryos may grow to larger sizes and indeed to the critical size.

Fisher (5) indicates that larger embryos are formed by a series of bimolecular reactions as shown below.

\[
\begin{align*}
&m \alpha_1 \rightleftharpoons \beta_m \\
&\beta_m + \alpha_1 \rightleftharpoons \beta_{m+1} \\
&\beta_{m+1} + \alpha_1 \rightleftharpoons \beta_{m+2} \\
&\beta_{m+2} + \alpha_1 \rightleftharpoons \beta_{m+3} \\
&\quad\vdots \\
&\text{etc.}
\end{align*}
\]

II.12
In this formulation \( m \) is the number of molecules in the small statistical groups above, \( \alpha_1 \) is a single molecule of the original \( \alpha \)-phase, and \( \beta_1 \) is an embryo of the new \( \beta \)-phase which contains \( i \) molecules. One might think of each different embryo size as a different chemical species. Then each of the equations in equation 11.12 can be thought of as a particular type of molecule reacting with molecules of \( \alpha \)-phase to produce a new type of molecule.

If a steady state system in which a number of different chemical species are reacting is visualized, it would be expected that after a period of time the concentrations of the various species would be adjusted such that the forward and reverse reaction rate are equal for each reaction. The system would then be in a state of equilibrium as long as no species is added to or removed from the system. An analogous situation is presumed to exist for the various sizes of embryos. An equilibrium number distribution will be produced after a certain period of time for the embryos containing \( i = m, m + 1, m + 2, \ldots, k - 1, k \) molecules where \( k \) is maximum number of molecules in any embryo. If \( n^* \) is the number of molecules in a nucleus for the conditions of the system under observation and if \( k \) is equal to \( n^* \), these embryos of \( \beta \)-phase or nuclei by definition could continue to grow and be observed as macroscopic vapor bubbles.

The general shape of the number distribution for
embryo sizes can inferred from the discussion up to this point. Referring to figure II.1 it is seen that any embryo with a size less than that of a nucleus will attempt to decrease the free energy of the system by collapsing to a smaller size. This means that in each of the growth steps of equation II.12 the reverse reaction predominates. Then for each of these reactions to be in equilibrium the concentration of "reactants" must be greater than the concentration of "products". This means there will be a continually decreasing number of embryos in each succeeding larger size of the growth process.

Fisher (5) indicates that this distribution would follow equation II.13.

\[ N_i \propto e^{-|\Delta F_i|/kT} \]  \hspace{1cm} \text{II.13}

In this equation \( N_i \) is the number density of embryos in the system at temperature \( T \) containing \( i \) molecules and \( \Delta F_i \) is the change in free energy required to form an embryo containing \( i \) molecules. Figure II.3 shows a typical number distribution along with the free energy change required for the formation of embryos containing \( i \) molecules. Depending on the conditions of the system, the distribution may or may not extend to \( i = n^* \), i.e. produce homogeneous nucleation.

If the number distribution of embryo sizes does indeed contain an embryo of size \( n^* \), this nucleus may con-
continue to grow and in effect be removed from the distribution and the system. The loss of this embryo will perturb the steady state, equilibrium number distribution in the same way as a chain of chemical reactions is perturbed if some of the products of the final reaction in the chain are removed from the system. In effect all reactions of the chain will act to restore the equilibrium concentrations by undergoing a net forward reaction. Then when a nucleus grows and effectively removes itself from the system, the reactions of equation II.12 will experience a net forward reaction to restore the lost embryo at the top of the distribution. However, this second nucleus
may also grow and leave the system and likewise all succeeding nuclei. In effect, then, a net forward reaction rate must take place in each reaction along the growth process. It can be assumed that the molecules leaving the system via the macroscopic bubbles are replaced in the system by an equal number of molecules of the original phase. This prevents a perturbation in the distribution from the bottom, i.e. a change in \( N_m \). This is the mechanism through which a homogeneous nucleation rate can be established. The succeeding treatment will deal with the conditions under which a reasonable nucleation rate will occur.

II.3 General Effect of Nuclear Radiation on the Homogeneous Nucleation Process.

When nuclear radiation interacts with a substance it may directly or indirectly deposit all or a portion of its energy in that substance. Types of radiation which deposit energy directly along their paths are fission fragments, alpha particles, and beta particles. Neutrons and gamma radiation do not deposit energy continuously along their paths but produce secondary energetic particles such as primary knock-on atoms and electrons. These particles then deposit along their paths the energy imparted to them by the original nuclear radiation. Much
of the energy which is deposited along an energetic particle's path is first transferred to the electronic structure of the substance through which it is passing. If the substance does not have a high electronic component of thermal conductivity, most of this deposited energy will finally manifest itself as molecular motion or thermal energy in a very small region about the energetic particle's path. This region may be on the order of 10 to 100 angstroms in diameter. In effect, then, the nuclear radiation or its secondary particles produce in a substance small regions containing highly energetic molecules. For a liquid system being subjected to radiation the existence of small regions containing molecules which are more like vapor than liquid molecules could be expected.

The above discussion suggests that the effect of nuclear radiation on the homogeneous nucleation process is to introduce a perturbation in the steady state number distribution of embryo sizes. This perturbation is a result of effectively feeding into the distribution an additional population of embryos of definite sizes from the radiation tracks. In order to simplify the physical picture it will be assumed that all the embryos from the radiation tracks are the same size.

Consider the reactions in the growth process which involves this embryo size, \( i = n_R \), from the radiation tracks. Since an additional number of embryos containing \( n_R \)
molecules now exists, a net forward reaction rate to \( n_{R+1} \) and a net reverse reaction rate to \( n_{R-1} \) will result. Assuming that the number of embryos coming from the radiation tracks is constant in time and that the maximum size embryo in the distribution does not contain the critical number of molecules, one would expect a new equilibrium number distribution to be established for \( i > n_R \) which is at a higher level than the corresponding distribution without radiation produced embryos. This higher distribution would make possible the creation of embryos with sizes greater than the maximum size in the process without radiation.

With the forward reactions for \( i > n_R \) in equilibrium, i.e. no net rate of reaction, there must be a net steady state reaction in the reverse direction for \( i < n_R \) if the concentration of embryos containing \( n_R \) molecules is to be constant or at an equilibrium level for a constant input rate of these embryos from the radiation tracks. An analogy can be drawn here between this situation and a water reservoir behind a dam in a river. If the river has a very low flow or no flow the water level will be just at the top of the dam and the reservoir will extend upstream.
a certain distance with ever decreasing water depth. Now if it rains steadily for a period of time creating a large steady flow in the river, the water level of the reservoir will have to rise in order to allow a greater flow over the dam and the increased level will extend the reservoir a greater distance upstream. Note that all the flow is downstream but the reservoir is longer. Likewise in this reaction chain all the $n_R$ embryos which are introduced must flow downstream or toward smaller sizes under equilibrium conditions. The resulting distribution would in general be as shown in figure II.4. The argu-

![Figure II.4 Effect of Radiation on the Number Distribution of Embryo Sizes](image)

ments made in the preceding section for the establishment of a nucleation rate should still apply in this case.
II.4 Detailed Treatment of the Homogeneous Nucleation Process with Radiation Produced Embryos

A system at a temperature, $T_v^* = T_1^*$, and pressure, $P_1$, will now be considered. The system is subjected to sufficient radiation such that the steady state population of embryos per unit volume containing $n_R$ molecules is $N_{n_R}$ and that a net forward reaction rate is occurring which produces $J$ nuclei per unit volume and per unit time in the system. Assuming steady state requires that each reaction in process from $i = n_R$ to $i = n^*$ have a net forward reaction equal to $J$. Note that asterisks refer to the state of the system which produces nuclei or critical sized embryo.

Consider any reaction in the chain such as $i = n$. Following Volmer (6) the net forward reaction in a time interval $dt$ per unit volume is the difference in the number of embryos of size $n$ which gain a molecule to become embryos of size $n + 1$ and the number of embryos of size $n + 1$ which lose a molecule to become embryos of size $n$ all per unit volume in a time interval $dt$. In other words the net forward rate of reaction per unit volume is the difference in the actual forward and reverse reaction rates per unit volume.

$$\beta_n + \alpha_1 \rightarrow \beta_{n+1}$$

$R(n \rightarrow n+1) =$ forward rate per unit volume

$$R(n \rightarrow n+1) = \text{forward rate per unit volume}$$
\[ R(n+1 \rightarrow n) = \text{reverse rate per unit volume} \quad \text{II.17} \]
\[ J(n \rightarrow n+1) = R(n \rightarrow n+1) - R(n+1 \rightarrow n) = J^* \quad \text{II.18} \]

The reaction rates can be expressed in the following manner. Let \( W(l \rightarrow v)(n) \) be defined as the number of molecules passing through the liquid-vapor interface from the liquid phase to the vapor embryo of \( n \) molecules per unit area and per unit time and \( A_n \) be the surface area of an embryo of \( n \) molecules. Then \( W(l \rightarrow v)(n) A_n dt \) is the probability that an embryo of \( n \) molecules will obtain one additional molecule in the time interval \( dt \). Likewise the probability that an embryo of \( n+1 \) molecules will lose a molecule in the time interval \( dt \) is \( W(v \rightarrow l)(n+1) A_{n+1} dt \). Here \( W(v \rightarrow l)(n+1) A_{n+1} dt \) is the flux of molecules passing through the interface from the vapor embryo to the liquid per unit time and per unit area and \( A_{n+1} \) is the surface area of an embryo of \( n+1 \) molecules. The reaction rates per unit volume are then equal to the probability of reaction per embryo times the number of embryos of that size per unit volume divided by the time interval \( dt \). If \( N_n \) is the number of embryos containing \( n \) molecules and \( N_{n+1} \) is the number of embryos containing \( n+1 \) molecules, the reaction rates of equations II.16 and II.17 are as follows.

\[ R(n \rightarrow n+1) = N_n \left[ W(l \rightarrow v)(n) A_n dt \right] / dt \quad \text{II.19} \]
\[ R(n+1 \rightarrow n) = \frac{N_{n+1} \left[ \bar{W}(v \rightarrow 1)(n+1)A_{n+1} dt \right]}{dt} \quad \text{II.20} \]

The net rate then becomes:

\[ J(n \rightarrow n+1) = N_n \bar{W}(1 \rightarrow v)(n)A_n - N_{n+1} \bar{W}(v \rightarrow 1)(n+1)A_{n+1} \quad \text{II.21} \]

Volmer (6) indicates that due to the finite size of a molecule crossing the interface of an embryo and the sharp curvature of the embryo surface, the embryo has an effective size which is exactly the same for a molecule crossing the interface in either direction. The effective radius of this surface is equal to the radius of an embryo of size \( n \) plus the radius of the molecule. Then:

\[ A_n = A_{n+1} = 4\pi \left( r_n + r_{\text{molecule}} \right)^2 = A_{\text{eff}}. \quad \text{II.22} \]

If the smallest embryo involved in this growth process, which is \( n_R \), is large compared to a single molecule, the effective area will be essentially equal to \( A_n \).

Equation II.21 can be simplified to the following:

\[ J(n \rightarrow n+1) \frac{A_n}{\bar{W}(v \rightarrow 1)(n+1)} = \frac{N_n \bar{W}(1 \rightarrow v)(n)}{\bar{W}(v \rightarrow 1)(n+1)} - N_{n+1} \quad \text{II.23} \]

Define \( \nu_{n+1} \) as the ratio of the forward reaction probability to the reverse reaction probability for the reaction being considered.
\[ \psi_{n+1} = \frac{\dot{W}(1\rightarrow v)(n) A_n}{\ddot{W}(v\rightarrow 1)(n+1) A_n dt} = \frac{\dot{W}(1\rightarrow v)(n)}{\ddot{W}(v\rightarrow 1)(n+1)} \quad \text{II.24} \]

\( \psi_{n+1} \) is also equal to the ratio of the flux of molecules into an embryo of size \( n \) to the flux of molecules out of an embryo of size \( n+1 \). Then equation II.23 becomes:

\[ \frac{J(n\rightarrow n+1) \psi_{n+1}}{A_n \dot{W}(1\rightarrow v)(n)} = N_n \psi_{n+1} - N_{n+1} \quad \text{II.25} \]

Equation II.25 is simply a convenient rearrangement of equation II.21. This equation applies to only one of the reactions of the reaction chain from \( i=n_R \) to \( i=n^* \). Similar expressions can be written for each of the other reactions in the chain as shown in equation II.26.

\[ \frac{J^*(n_R\rightarrow n_R+1) \psi_{n_R+1}}{A_{n_R} \dot{W}(1\rightarrow v)(n_R)} = N_{n_R} \psi_{n_R+1} - N_{n_R+1} \quad \text{II.26} \]

\[ \frac{J^*(n_R+1\rightarrow n_R+2) \psi_{n_R+2}}{A_{n_R+1} \dot{W}(1\rightarrow v)(n_R+1)} = N_{n_R+1} \psi_{n_R+2} - N_{n_R+2} \]

\[ \vdots \]

\[ \frac{J^*(n^*-1\rightarrow n^*) \psi_{n^*}}{A_{n^*-1} \dot{W}(1\rightarrow v)(n^*-1)} = N_{n^*-1} \psi_{n^*} - N_{n^*} \]
As stated above the net reaction rates for all reactions in the chain are equal for a steady state nucleation rate. Also appearing on the left hand side of each equation of equation II.26 is the flux of molecules from the liquid to the different sized embryos. It will be assumed that this flux is the same for all the embryos regardless of size since it depends mainly on the liquid conditions which are the same around all embryos. Then the quantity \( J^*/W(1\to v) \) can be taken as constant for this entire set of expressions.

If one assumes that all the reaction probabilities or fluxes and, therefore, \( \psi \)'s can be calculated, then the only unknowns in the set of equations II.26 are the nucleation rate and the number densities of the different sized embryos. It can be assumed that the number density of embryos at the starting point of the overall forward reaction, \( N_{n_R} \) at \( n_R \), is known. It was indicated in a preceding discussion that the nuclei could be assumed to grow and in effect leave the system. Therefore, \( N_{n^*} \) can be taken as zero. Then the unknowns consist of \( N_{n_R+1}, \ N_{n_R+2}, \ldots, N_{n^{*-1}} \) and \( J^* \) which number \( (n^{*-} - n_R) \) in all. Note that equation II.26 consists of \( (n^{*-} - n_R) \) expressions so that in theory all unknowns can be obtained. There is little interest in knowing the number densities at the intermediate embryo sizes so a mathematical procedure will be used which eliminates these quantities and produces a solution for the nucleation rate, \( J^* \).
The procedure involves dividing each of the expressions in equation II.26 by the product of the $\psi$'s from $\psi_{n+1}$ to the $\psi$ appearing in the expression. The result for the general reaction is shown in equations II.27 and II.28.

\[
\frac{J \psi_{n+1}}{A_n^W(k \rightarrow v)} = N_n \psi_{n+1} - N_{n+1} \text{ all multiplied by II.27}
\]

\[
\frac{1}{\psi_{n+1} \cdots \psi_{n+1} \cdots \psi_{n+1}} = 1 \prod_{i=n+1}^{n+1} \psi_i
\]

gives:

\[
\frac{J}{A_n^W(1 \rightarrow v) \psi_{n+1} \cdots \psi_n} = \frac{N_n}{\psi_{n+1} \cdots \psi_{n+1}} - \frac{N_{n+1}}{\psi_{n+1} \cdots \psi_{n+1} \psi_{n+1}}
\]

\[
= N_n \prod_{i=n+1}^{n+1} \psi_i - N_{n+1} \prod_{i=n+1}^{n+1} \psi_i \text{ II.28}
\]

Applying this procedure to equation II.26 gives:

\[
\frac{J^*}{A_n^W(1 \rightarrow v)} = N_n \psi_{n+1} - \frac{N_{n+1}}{\psi_{n+1} \psi_{n+1}} \text{ II.29}
\]

\[
\frac{J^*}{A_n^W(1 \rightarrow v) \psi_{n+1} \cdots \psi_{n+1} \psi_{n+1}} = \frac{N_{n+1} + 1}{\psi_{n+1} \psi_{n+1}} - \frac{N_{n+2}}{\psi_{n+1} \psi_{n+1} \psi_{n+2}}
\]
\[
\frac{J^*}{A_{n_R+2}W(1\rightarrow v)} \frac{\psi_{n_R+1} \psi_{n_R+2}}{\psi_{n_R+1} \psi_{n_R+2}} = \frac{N_{n_R+2}}{\psi_{n_R+1} \psi_{n_R+2}} - \frac{N_{n_R+3}}{\psi_{n_R+1} \psi_{n_R+2} \psi_{n_R+3}}
\]

\[\vdots\]

\[
\frac{J^*}{A_{n^*-1}W(1\rightarrow v)} \frac{\psi_{n_R+1} \ldots \psi_{n^*-1}}{\psi_{n_R+1} \ldots \psi_{n^*-1}} = \frac{N_{n^*-1}}{\psi_{n_R+1} \ldots \psi_{n^*-1}} - 0
\]

Now when this set of equations is added together the intermediate terms on the right hand side cancel out resulting in:

\[
\frac{J^*}{W(1\rightarrow v)} \left[ \frac{1}{A_{n_R}} + \frac{1}{A_{n_R+1} \psi_{n_R+1}} + \frac{1}{A_{n_R+2} \psi_{n_R+1} \psi_{n_R+2}} + \cdots + \frac{1}{A_{n^*-1} \psi_{n_R+1} \ldots \psi_{n^*-1}} \right] = \frac{N_{n_R}}{n_R}
\]

or:

\[
\frac{J^*}{W(1\rightarrow v)} \sum_{n=n_R}^{n=n^*-1} \left[ A_n \prod_{i=n_R+1}^{n} \psi_i \right]^{-1} = \frac{N_{n_R}}{n_R}
\]

This equation gives the rate at which critical nuclei are formed per unit time and volume for a certain number density of initial embryos of size \(n_R\). The problem of defining the initial embryos as to size and number and the problem of determining the other quantities in equation
II.31 still must be handled.

It was assumed above that the fluxes of molecules across the embryo interfaces were known quantities. Expressions for these fluxes and for the $\psi$'s will now be obtained. If one considers the vapor as a perfect gas, an expression for the flux can be calculated assuming a Maxwellian distribution of molecules and that the velocity distribution is isotropic in direction. Kennard (7) gives the following expression.

$$W(v \rightarrow 1) = \frac{1}{4\sqrt{\pi N\bar{v}\bar{v}}} \frac{\alpha}{P_v} = \frac{\alpha}{\left[2\pi mkT\right]^{1/2}}$$

II.32

where $N$ is the molecule density, $\bar{v}$ is the mean molecular speed, $\alpha$ is the condensation coefficient, $P_v$ is the pressure of the vapor, $m$ is the mass of a molecule, $k$ is Boltzmann's constant, and $T$ is the absolute temperature. The flux from the liquid to the vapor can be obtained by considering a liquid in equilibrium with its vapor. In this situation the rate of molecules leaving the liquid to become vapor is just equal to the number of molecules returning from the vapor to the liquid. Therefore:

$$W(1 \rightarrow v)_{\text{Equilibrium}} = W(v \rightarrow 1)_{\text{Equilibrium}} = \frac{\alpha}{\left[2\pi mkT\right]^{1/2}}$$

II.33

where $P_\infty$ refers to the equilibrium pressure for a flat
interface or infinite radius.

Assuming the flux of molecules into an embryo is independent of the size of the embryo, it can be evaluated for any particular embryo which is desirable. An embryo of critical size is assumed to contain vapor which is in equilibrium with the surrounding liquid, i.e. the rates of molecules leaving and entering the critical embryo are exactly equal. Note that evaluation of $W(1\rightarrow v)$ under these conditions may be slightly in error since a curved interface is involved instead of a flat interface.

$$W^*(1\rightarrow v) = \frac{\bar{c}}{\sqrt{2\pi mkT^*}} \approx W(1\rightarrow v)_{all embryos} \quad \text{II.34}$$

The assumption that $W(1\rightarrow v)$ is constant for all embryos should perhaps be justified at this point. This can be done by looking at the actual evaporation process on a molecular scale as indicated by Loeb (8). The analysis of this process in effect integrates over the energy distribution of molecules in the liquid phase to find the molecules which are capable of overcoming the energy barrier against evaporation. In the case of a flat interface this barrier is essentially the work that must be done by a molecule against the Van der Waal's or intermolecular forces in the liquid and the surface forces in order to escape from the liquid. This work is just the heat of vaporization per molecule for the flat interface.
For the case of evaporation into an embryo considered in this work there is an additional energy barrier, the free energy change associated with the growth of the embryo. If a molecule of liquid desired to become part of an embryo of \( i \) molecules thus giving the embryo \( i+1 \) molecules, the free energy of the embryo has changed by the amount given in equation II.35 where \( \Delta F(1) \) is the free energy of formation of an embryo of \( i \) molecules. If it is assumed that the pressure in the embryo can be related to the liquid pressure through the dynamic equilibrium expression, equation II.8 for the free energy of formation of an embryo of mass \( m \) becomes:

\[
\Delta F = -m_v (\mu_v - \mu_1) - \frac{1}{2} \sigma A
\]

Then, by defining the average chemical potentials per molecule, where \( M \) is the molecular weight and \( N_Av \) is Avagadro's number, as \( \phi = \mu M / N_Av \), equation II.36 becomes:

\[
\Delta F_1 = -1 \left\{ \phi_v \left[ \frac{T}{P} \right] - \phi_1 (T, P_1) \right\} - \frac{1}{2} \sigma A_1
\]

Equation II.35 can then be written as:
This assumes that \((P_v)_{i+1} \approx (P_v)_i\) and, therefore, 
\[ W_{i \rightarrow i+1} = -(i+1) \phi_v \left[ T, (P_v)_{i+1} \right] + i \phi_v \left[ T, (P_v)_i \right] + \phi_1(T, P_1) - \frac{1}{3} \sigma (A_{i+1} - A_i) \]

\[ W_{i \rightarrow i+1} \approx \phi_v \left[ T, (P_v)_i \right] - \phi_1(T, P_1) \]  

This assumes that \((P_v)_{i+1} \approx (P_v)_i\) and, therefore, 
\[ \phi_v \left[ T, (P_v)_{i+1} \right] \approx \phi_v \left[ T, (P_v)_i \right] \] and that \(A_{i+1} \approx A_i\) which is probably fairly good since the addition of one molecule is a very small change. Then, a molecule in the liquid must not only be sufficiently energetic to overcome the heat of vaporization but also the change in chemical potential that may exist. It can be seen that the chemical potential of the vapor in the embryo is a function of the embryo vapor pressure and therefore the embryo size. It is this term which makes the flux of molecules into embryos of different sizes unequal. Then, it must be concluded that the assumption of constant \(W_{(l \rightarrow v)}\) requires that \(W_{i \rightarrow i+1} \ll \lambda\) where \(\lambda\) is the heat of vaporization per molecule. This situation does exist in the range of conditions of interest in this work, i.e. far away from the critical point.

The flux from a vapor embryo of \(i\) molecules at the same temperature, since this growth process is an isothermal process is:

\[ W_{(v \rightarrow l)}(i) = \frac{\alpha (P_v)_i}{2 \pi mkT^*} \]  

II.39
where \((P_v)_i\) is the vapor pressure in an embryo of \(i\) molecules and is related to the embryo size through the dynamic equilibrium criterion for spherical embryos.

\[
(P_v)_i = P_1 + \frac{2\sigma}{r_i} \tag{II.40}
\]

In this equation \(r_i\) is the radius of an embryo of \(i\) molecules. Combining these last three equations with the definition of \(\psi_i\) gives:

\[
\psi_i = \frac{\bar{W}(i\rightarrow v)}{W(v\rightarrow 1)(1)} = \frac{P^*_v}{P_1 + 2\sigma/r_i} \tag{II.41}
\]

The product of the \(\psi\)'s from \(i=n_i\) to \(i=n\) will be calculated again following Volmer(6). By using the fact the exponential of the natural logarithm of a number is equal to the number itself, the product of the \(\psi\)'s will equal the product of exponentials and consequently an exponential with a summation for an exponent.

\[
\prod_{i=n_R+1}^{n} \psi_i = \prod_{i=n_R+1}^{n} e^{\ln \psi_i} = \exp \left[ \sum_{i=n_R+1}^{n} \ln \psi_i \right] \tag{II.42}
\]

Since the addition of one molecule is such a small step change in the logarithm term in equation II.42, the summation can be replaced by an integral without introducing appreciable error. Then equation II.42 becomes:
\[
\prod_{i=n_{R}+1}^{n} \psi_{i} = \exp \left[ \int_{i=n_{R}}^{n} \ln \psi_{i} \, di \right] \quad \text{II.43}
\]

Note that the lower limit on the integral is changed to give a non-zero value for \( \psi_{n_{R}+1} \).

This integral can be evaluated after suitable manipulation. The expression for \( \psi_{i} \) or \( \psi(1) \) is given by equation II.41. The definitions shown in equations II.44 and II.45 along with equation II.40 are introduced into equation II.41 to give equation II.46.

\[
b = \left( \frac{P_{*}}{P_{v}} - P_{l} \right) / P_{*} \quad \text{II.44}
\]

\[
x(i) = \frac{r(i)}{r*} \quad \text{II.45}
\]

\[
\frac{1}{\psi(1)} = \frac{P_{l} + 2\sigma/r(i)}{P_{*}} = 1 - b + b/x(i) \quad \text{II.46}
\]

Before the integral in equation II.43 can be carried out a relationship between \( i \) and \( x \) must be found. This relationship can be obtained from the perfect gas law.

\[
P_{v}(i) V(i) = i k T* \quad \text{II.47}
\]

Here \( V(i) \) is the volume of an embryo of \( i \) molecules and is equal to \((4/3)\pi r(i)^{3}\). \( P_{v}(i) \) is the pressure in this embryo.
Forming the ratio of this equation and one like it for a critical embryo also at $T^*$ gives, using equation II.46:

$$\frac{P_v(i) V(i)}{P^*_v V^*} = \frac{(4/3) \pi r(1)^3 \left[ P_1 + 2 \sigma / r(1) \right]}{(4/3) \pi r^* 3 P^*_v} = \frac{x(i)^3 \left[ P_1 + \frac{2 \sigma}{r(1)} \right]}{P^*_v}$$

$$= x(i)^3 \left[ 1 - b + \frac{b}{x(i)} \right] = 1/n^* \quad \text{II.48}$$

Then:

$$d_1 = n^* \left[ 3(1 - b)x(i)^2 + 2bx(i) \right] dx \quad \text{II.49}$$

Even though the perfect gas law does not apply well to saturated vapors, the error introduced here should tend to cancel out because a ratio is used. The limits on the integral are $x(n)$ and $x(n_R)$ as defined by equation II.48 at $i=n$ and $i=n_R$, respectively. The negative sign comes about because the argument of the logarithm was inverted.

\[ \prod_{i=n_R+1}^{n} \psi_1 = \exp \left\{ -n^* \int_{x(n_R)}^{x(n)} \left[ \ln(1-b+b/x) \right] \left[ 3(1-b)x^2 + 2bx \right] dx \right\} \quad \text{II.50} \]

The integral exponent becomes:

$$-n^* \int_{x(n_R)}^{x(n)} \left[ \ln(1-b+b/x) \right] \left[ 3(1-b)x^2 + 2bx \right] dx \quad \text{II.51}$$
\[
= -n^* \left\{ x(n)^3 \left[ 1-b+b/x(n) \right] \ln \left[ 1-b+b/x(n) \right] + \frac{b}{2} x(n)^2 \right\} \\
+ n^* \left\{ x(n_R)^3 \left[ 1-b+b/x(n_R) \right] \ln \left[ 1-b+b/x(n_R) \right] + \frac{b}{2} x(n_R)^2 \right\}
\]

Going back to equation II.37 for the free energy of formation of an embryo of \( i \) molecules in terms of the quantities in equation II.51 will give some physical insight into this up-to-now mathematical procedure.

Consider first the thermodynamic relation for the change in chemical potential with pressure at constant temperature for a single component substance from Hatsopoulous (3).

\[
\left( \frac{\partial Z}{\partial P} \right)_T = V
\]

or

\[
\left( \frac{\partial Z}{\partial P} \right)_T = \left( \frac{\partial \mu}{\partial P} \right)_T = N_A \left( \frac{\partial \phi}{\partial P} \right)_T = v \quad \text{II.52}
\]

Then, using the gas law for the vapor in the embryos the average chemical potential per molecule in the embryo can be obtained as a function of pressure since it is being assumed that the entire distribution of embryos is isothermal. At the critical size the chemical potentials of

\[
N_A \int d\phi_v = RT \int_{P_v}^{P_v^*} \frac{dP_v}{P_v}
\]

\[
\phi(P_v) - \phi(P_v^*) = kT \ln(P_v/P_v^*) \quad \text{II.53}
\]
the vapor and liquid molecules are equal. Then, equation II.53 can be expressed as:

\[ \psi_v(T, P_v(1)) - \psi_1(T, P_1) = kT \ln \left[ \frac{P_v(1)}{P_v^*} \right] \quad \text{II.54} \]

and equation II.37 becomes:

\[ \Delta F(i) = -kT \ln \left[ \frac{P_v(i)}{P_v^*} \right] - \frac{1}{3} \sigma^4 \pi r(i)^3 \quad \text{II.55} \]

Then using equations II.44, II.45, II.46, and II.47 this expression becomes:

\[ \frac{\Delta F(i)}{kT} = -n^* \left\{ x(i)^3 \left[ 1 - b + b/x(i) \right] \ln \left[ 1 - b + b/x(i) \right] + \frac{b}{2} x(i)^2 \right\} \quad \text{II.56} \]

Notice now the similarity of this expression and equation II.51. Making the obvious substitution into equation II.51, equation II.50 becomes:

\[ \sum_{i=n_R+1}^{n} \psi_i = \exp \left[ \frac{\Delta F(n)}{kT} - \frac{\Delta F(n_R)}{kT} \right] \quad \text{II.57} \]

\( \psi(1) \) was defined in equation II.41 as a ratio of the flux of molecules into an embryo to the flux of molecules out of an embryo containing \( i \) molecules. This ratio can also be thought of as the relative probability that an
embryo of \( i \) molecules will grow to one of \( i+1 \) molecules. Then, the product of these relative probabilities for each step of the growth process can be thought of as the relative probability of embryo growth from one size to another. Equation II.57 indicates that the relative probability of an embryo of size \( n_R+1 \) reaching a size \( n \) is a function of the difference in the free energies of formation of these two sizes. The free energies of formation in equation II.57 are negative and \( \Delta F(n) > \Delta F(n_R) \) for \( n > n_R \). The exponent is then negative and its magnitude increases for the same \( n_R \) as \( n \) increases. This simply says that the probability of growth to a particular size decreases as the size increases. Note the similarity between equations II.57 and II.13 which gives the number distribution for the case of \( n_R=0 \). This similarity should be expected since the population of a particular size of embryo should be proportional to the probability of growth to that size.

Looking now at equation II.31, the summation over \( n \) can be replaced with an integral also without appreciable error. If \( A_n \) can be put in terms of \( x(n) \), the integrand will be a function of \( x(n) \) only.

\[
A_n = 4\pi r_n^2 = 4\pi r^*^2(r_n^2/r^*^2) = A*x(n)^2 = A(n) \quad \text{II.58}
\]

Then solving for \( J* \) in equation II.31, substituting for \( W(1 \rightarrow v) \) from equation II.34, \( A(n) \) from equation II.58, the
product of the $\psi$'s from equations II.57 and II.56, and $dn$ from equation II.49 where $i=n$, the equation for the nucleation rate per unit volume is:

$$J^* = \frac{A*N_{nR} \propto P^*}{n^* \sqrt{2\pi m k T^*}} \left[ \int_{x(n_R)} \left[ 3(1-b) + 2b/x(n) \right] \exp \left[ \frac{\Delta F(n_R)}{kT^*} \right] \right]^{-1} \exp \left\{ -\frac{\Delta F[x(n)]}{kT^*} \right\} dx(n)$$

II.59

$$J^* = \frac{A*N_{nR} \propto P^*}{n^* \sqrt{2\pi m k T^*}} \exp \left[ -\frac{\Delta F(n_R)}{kT^*} \right] \frac{1}{I_1(n_R)}$$

II.60

where $I_1(n_R)$ is defined as the integral given in equation II.61.

$$I_1(n_R) = \int_{x(n_R)} \left[ 3(1-b) + 2b/x(n) \right] \exp \left[ n^* x(n) \right] \left[ 1-b+b/x(n) \right]$$

(times) $\ln \left[ 1-b+b/x(n) \right] + \frac{n^* b}{2} x(n)^2 dx(n)$

II.61

II.5 Determination of the Embryo Density from the Radiation Tracks

The nucleation rate is now defined in terms of the embryo size coming from the radiation tracks, the state of
the system under consideration, and the population of embryos per unit volume in the system from the radiation tracks, i.e. \( N_{n_R} \). This last term is required in a more definitive form. Since its magnitude is related to the number distribution which is the basis of the above development, it will be treated next.

As indicated previously the rate of embryo input to the system at size \( n_R \) must equal the net reverse reaction rate in the number distribution from \( i=n_R \) to essentially \( i=0 \) under steady state conditions and no nucleation. If it is assumed that the nucleation rate is small compared to the embryo production rate in the radiation tracks, the net reverse reaction rate will very nearly equal the embryo production rate in the tracks even when nucleation is taking place. Then the number density of these embryos from the radiation tracks must be such that the necessary reverse reaction is possible. This net reverse reaction rate must be:

\[
J^- = \text{Production Rate of } n_R \text{ Embryos} = R(n_R) \quad \text{II.62}
\]

If precisely the same procedure is employed as in the preceding section to find the nucleation rate, \( J^- \) can be calculated. A set of equations analogous to equation II.29 can be set up from \( i=m \) to \( i=n_R \) where \( m \) is the size of the smallest embryos in the distribution. In this case,
however, the number distribution will be assumed to reduce to the number distribution without radiation at very small embryo sizes, i.e. the radiation produced embryos completely condense. When each equation of this new set is multiplied by the appropriate product of \( \psi \)'s and the set is added together, equation II.63 results which is analogous to equation II.30.

\[
\frac{J^{-}}{W(1 \rightarrow v)} \left[ \frac{1}{A_m} + \frac{1}{A_{m+1} \psi_{m+1}} + \frac{1}{A_{m+2} \psi_{m+1} \psi_{m+2}} + \cdots + \right]
\]

\[\text{II.63}\]

\[
\frac{1}{A_{n_R-1} \psi_{m+1} \psi_{m+2} \cdots \psi_{n_R-1}} = -N_{n_R} \prod_{i=m+1}^{n_R} \psi_i + N_m
\]

Note that this equation results directly from equation II.30 if \( n_R \) is replaced by \( m \), \( N_{n_R} \) is replaced \( N_m \), and \( N_{n_R}^* \) is replaced by \( N_{n_R} \). Continuing the analogy it can be seen that the product of the \( \psi \)'s is given by:

\[
\prod_{i=m+1}^{n} \psi_i = \exp \left[ \frac{\Delta F(n)}{kT^*} \right]
\]

\[\text{II.64}\]

Again the exponent is defined by equation II.56. Then equation II.63 can be put in integral form as before to give:
\[
\frac{J^{-}}{W(1 \rightarrow v)} \int_{x(m)}^{x(n_{R})} \frac{n^{*} [3(1-b)+2b/x(n)] dx(n)}{A^{*} \exp \left[ \frac{\Delta F[x(n)]}{kT} \right]} \\
= -N_{n_{R}} \exp \left[ -\frac{\Delta F(n_{R})}{kT} \right] + N_{m}
\]

Rearranging and substituting for \( W(1 \rightarrow v) \) gives:

\[
N_{n_{R}} = -\frac{J^{-} n^{*} \sqrt{2 \pi m kT^{*}}}{\alpha} \frac{\exp \left[ \frac{\Delta F(n_{R})}{kT^{*}} \right]}{A^{*} \frac{\Delta F[x(n)]}{kT^{*}}} \\
\times (\text{times}) \int_{x(m)}^{x(n_{R})} \left[ 3(1-b)+2b/x(n) \right] \exp \left[ -\frac{\Delta F[x(n)]}{kT^{*}} \right] dx(n) \\
+ N_{m} \exp \left[ \frac{\Delta F(n_{R})}{kT^{*}} \right]
\]

\[
= -\frac{J^{-} n^{*} \sqrt{2 \pi m kT^{*}}}{\alpha} \frac{\exp \left[ \frac{\Delta F(n_{R})}{kT^{*}} \right]}{A^{*} \frac{\Delta F[x(n)]}{kT^{*}}} I_{2}(n_{R}) + N_{m} \exp \left[ \frac{\Delta F(n_{R})}{kT^{*}} \right]
\]

where \( I_{2}(n_{R}) \) is defined as the integral given in equation II.67.

\[
I_{2}(n_{R}) = \int_{x(m)}^{x(n_{R})} \text{Integrand of } I_{1}(n_{R}) \ dx(n) \quad \text{II.67}
\]
The minus sign in equation II.66 results from the definition of the net forward reaction as positive in equation II.18. In this case $J^-$ is, therefore, negative which gives a positive value of $N_{n_R}$.

Equation II.66 for the required number density of embryos of the size produced in the radiation tracks indicates that the number density must increase for a constant production rate as the size of these embryos approach the size of a critical embryo. This trend has a physical basis in that the tendency for an embryo to collapse near the critical size is much less than for a smaller embryo. Therefore, since the probability of decreasing in size is smaller for larger embryos, the population must be larger in order to maintain a constant reverse reaction. Note also that when $J^-$ is zero, i.e. no radiation present, the number density at $n_R$ is given by an equation like II.13 as indeed it should.

Combining equations II.60 and II.66 gives the final equation for the nucleation rate per unit volume. This equation is a function of the size of the embryos produced in the radiation tracks and the rate of embryo production per unit volume from the tracks. The size of the embryos produced will be considered in the next chapter. The rate of embryo production per unit volume is a function of the intensity of the radiation interacting with the system and the number of embryos which result from each track. The
number of embryos per track will also depend on the size of the embryos produced so for the purposes of this general theory the number per track will be defined as $X(n_R)$. Then defining the radiation reaction rate per unit volume as $\text{RRR}$, the embryo production rate per unit volume becomes:

$$ R(n_R) = X(n_R) \cdot \text{RRR} \quad \text{II.69} $$

And equation II.68 becomes:

$$ J^*(n_R, \text{RRR}) = X(n_R) \cdot \text{RRR} \cdot \frac{I_2(n_R)}{I_1(n_R)} + \frac{N_m A^* \alpha P^*}{n^* \sqrt{2\pi m k T^*}} \cdot \frac{1}{I_1(n_R)} \quad \text{II.70} $$

In equation II.70 the term $N_m$ has not been defined quantitatively. At the beginning of this chapter mention was made of the assumed existence of small statistical groups of $m$ molecules each which could be thought of as the smallest vapor embryos in the number distribution. $N_m$ is the density of these embryos. Volmer (6) indicates that this density should be a function of the heat of vaporization per molecule, $\lambda$, and the total molecule density in the liquid, $N_1$, as shown in equation II.71.

$$ N_m = N_1 e^{-\lambda/kT^*} \quad \text{II.71} $$
It is interesting to take a qualitative look at equation II.70. Note what happens if the radiation produced embryos cease to be produced, i.e. $\Delta R = 0$. The first term on the right hand side goes to zero and the remaining term can be shown to be equivalent to the expression obtained by Volmer (6) for pure homogeneous nucleation free of outside influences. Also as the size of the embryos produced in the radiation tracks tends toward $n_R \rightarrow m$, $I_2(n_R)$ tends toward zero and again the expression for pure homogeneous nucleation results as indeed it should. Then, finally, looking at the opposite limit, i.e. $n_R \rightarrow n^*$, $I_1(n_R)$ tends toward zero and the nucleation rate tends to infinity. In actual fact this does not occur but results in this development due to the integral representation of the process when only a few steps in the growth process are involved. The fact that the nucleation rate increases greatly at this point indicates independence of the homogeneous growth process. The embryos produced in the tracks are already of critical size. Note also that the assumption made in the development of $N$ that a steady reverse reaction rate is set up which is equal to the production rate of embryos in the radiation tracks begins to break down at this point since a significant portion of these radiation produced embryos now participate in the forward reaction.

II.6 Criterion for the Existence of Radiation Induced Homogeneous Nucleation
The development of the above theory has led to a homogeneous nucleation rate per unit volume for a certain type of radiation interacting at a certain rate with a system at a prescribed state. For the purposes of this thesis it is desired to determine the threshold state of the system for which radiation induced nucleation will occur. Equation 11.70 gives a rate of nucleation per unit volume and cannot predict the threshold conditions until something is said about the system variables of radiation reaction rate, system volume, and observation time. Nucleation would be more likely to occur in a very large system which is under observation for a long period of time and subjected to an intense source of radiation.

The criterion for the system state for which radiation induced nucleation will be said to occur shall be the formation of one nucleus in a system of volume $V_s$ in an observation time $T$ during which the system is exposed to a radiation reaction rate $RRR$ and during which the system state is held constant. Then if the rate of nucleation as given by equation II.70 is multiplied by the system volume and observation time the resulting number must be unity to satisfy the criterion. The criterion in equation form is given by equation II.72. The application of this criterion to a particular system and the comparison of the predicted results with experimental data will be reserved for later chapters.
\[ x(n_R) \text{ RRR } v_s \frac{I_2(n_R)}{I_1(n_R)} + \frac{V_s \tau N_1 e^{-\lambda/kT^*}}{n^* \sqrt{2\pi mkT^*}} - \frac{A^* \alpha^{p*}}{v} \frac{1}{I_1(n_R)} = 1 \]
III.1 Required Energy of Formation of an Embryo

It was determined in the previous chapter that the
formation of an embryo in a system at a pressure $P_1$ and a temperature $T_1^* = T_v^*$ involved a change in free energy. This change in free energy or the work of formation for an embryo of size $n$ was given by equation II.56.

Another energy requirement is the vaporization energy involved in changing the phase of the material in the nucleus. In the previous chapter this energy did not enter the treatment because the embryo growth was brought about by the addition of molecules from the liquid which were sufficiently energetic to more than overcome the intermolecular bonding in the liquid. Then, the embryo in a sense was selectively feeding on the high energy molecules of the liquid, thus obtaining during its growth not only the necessary heat of vaporization but also the necessary free energy of formation from the superheated liquid. In the present analysis, however, the embryo is assumed to be produced solely by an external influence, i.e. radiation. This energy of vaporization or more accurately the change in enthalpy from superheated liquid at $T_1 = T_v$ and $P_1$ to vapor at $T_v$ and $P_v$ will be to a good approximation equal to the heat of vaporization per unit mass, $h_{fg}$ which is evaluated at $T_v$, times the mass of vapor in the embryo.

Since the energy of the radiation is deposited in a cylindrical region less than 100 A in diameter, this region could be expected to be very energetic with an associated high temperature and pressure. This small
region would also be expected to expand rapidly and in doing so would impart a certain kinetic energy to the surrounding liquid. In the final stages of this expansion process, however, the embryo will in effect be expanded by the inertia of the moving liquid as it slows down. The expansion will be concluded when and only when all the kinetic energy of the liquid has been dissipated and the system is static. If the embryo at this point is not critical a collapse mechanism will begin. In effect, then, the only energy actually lost in this process is that due to conduction away from the vapor region and the viscous dissipation due to the movement of a viscous fluid. These two effects will be considered in the next section.

In addition to the three energy requirements mentioned above there may be energy sinks in particular substances, such as the dissociation of water in the radiation track to form hydrogen and oxygen gas. This formation of gas will affect not only the energy available for formation, $E_A$, but also the internal pressure of the embryo. These effects will be treated in the next chapter when a specific case is treated.

The total energy of formation for an embryo of size $n$ is, then, given in qualitative terms by equation III.1 where $\Delta F(n)$ is the total free energy change for the formation of an embryo of size $n$. 
\[ E_{\text{formation}}(n) = \Delta F(n) + \Delta H(n)_{\text{vaporization}} + \Delta E(n)_{\text{expansion losses}} \]

III.2  Energy Losses Associated with the Expansion of the Radiation Track

Seitz (9) indicates that the criterion for the occurrence of radiation induced nucleation may not be the satisfaction of the energy requirements of equation III.1, but the completion of the process in a critical time period such that appreciable energy is not conducted away from the hot vapor region. He suggests that it may be necessary to require more energy in the track to force the expansion process to take place in a sufficiently short time period. Analytically this is a problem of solving the dynamics equations to find the velocity of expansion and the total time for expansion to the critical size for a particular initial pressure and energy in the radiation track. Then, the actual energy of formation becomes the energy which produces an initial pressure in the track such that expansion to critical size will take place within the critical time period.

Norman (10) uses this same basic criterion of a critical expansion time to provide a required average expansion velocity for the process. From this a liquid kinetic energy is obtained which is assumed to be lost
to the process. Then, an equation like III.1 gives the total energy of formation of the nucleus.

The basic assumption underlying these two approaches is that the hot radiation track can be treated as a quenching phenomenon in which a material at high temperature is suddenly immersed in a relatively cold bath. The relaxation time, which is the time interval for the energy contained in the hot material to decrease by a factor of $e$, is taken as the effective lifetime for the expansion process. This relaxation time is given by equation III.2 where $r$ is the radius of expanding region and $D$ is the thermal diffusivity. Then, if the distance over which the expansion takes place is divided by this time, an approximate average velocity for expansion results. Using this average interface velocity one can establish a velocity as a function of distance throughout the liquid and integrate over all the liquid to find the kinetic energy contained in the liquid. This method gives the kinetic energy as:

$$K.E. = \rho_1 D^2 r$$  \hspace{1cm} III.3

It turns out that for water this method gives a kinetic
energy which is small compared with the other terms which make up the total energy of formation. However, for a different substance such as sodium the thermal diffusivity is greater than that for water by more than two orders of magnitude. This means that the kinetic energy imparted to the liquid sodium would be four or five orders of magnitude greater than for water and it would be by far the dominant term in the energy of formation. The Seitz approach would require expansion velocities on the order of several hundred times greater than for water and, indeed, sonic velocity consideration could make it nearly impossible to have any appreciable expansion at all. It would appear that this basic assumption may not be sufficiently valid to be generally applicable.

The basic problem in the above approach is the assumption that the energy in the initial radiation track may be lost in the same way that energy is lost from a hot piece of metal when put into a cold bath. Since evaporation takes place during the expansion and energy is "soaked up" in the evaporation process at the interface during expansion, it seems possible that the energy attempting to leave the vapor region through the interface might well be returned to the region in the form of heat of vaporization of the mass being added to the region. The fact that additional mass must be added to the vapor region can be established by comparing the mass of liquid
in the region of initial energy deposition which can be assumed to be completely converted to vapor and the final mass of vapor in the track at the completion of expansion. Then, since there is evaporation at the interface and since the interface must act as a heat sink, a reasonable assumption could be that little or no energy escapes during the expansion process.

Then, if the energy loss problem can be neglected, the dynamics of the process can be relaxed to a "minimum energy" expansion in which nearly all of the energy imparted to the liquid as kinetic energy can be recouped. The process now has sufficient time to proceed to the static state, thereby, incurring only viscous losses as a result of expansion. Seitz (9) indicates that viscosity is probably important in the dynamics of the expansion for fluids with viscosities of one centipoise or greater. Then, it is probably reasonable to assume that viscous dissipation in the liquid can be neglected for fluid with viscosities substantially less than one centipoise.

On the basis of the above discussion the energy requirement for expansion losses, i.e. conduction losses and viscous losses, will be neglected.

III.3 General Equation for the Energy of Formation of an Embryo

Substituting equation II.56 into equation III.1 and
expressing the volume of the embryo in terms of the

dimensionless size $x$ defined by equation II.45 gives:

\[
E_f(n) = n * kT * \left\{ x^3(n) \left[ 1 - b + b / x(n) \right] \ln \left[ 1 - b + b / x(n) \right] + \frac{b}{2} x(n)^2 \right\} +
\]

\[
\frac{4}{3} \pi x^3(n) r^3 \rho_v h_{fg}
\]

Equation III.4 can be further simplified with the gas

law since $n * kT = P_v V^* = P_v (4/3) \pi r^3$. Then, in effect

since $b$, $\rho_v$, and $h_{fg}$ are functions of the state of the

system, $T_v^* = T_1$ and $P_1$, the energy of formation of an

embryo of radius $r_n$ containing $n$ molecules is also a

function of the system temperature and pressure. Then,

equation III.4 becomes:

\[
E_f(n, T_1, P_1, \text{Substance}) =
\]

\[
\frac{4}{3} \pi r^3 x(n)^3 \left\{ P_v \left[ 1 - b + b / x(n) \right] \ln \left[ 1 - b + b / x(n) \right] + \frac{P_v b}{2} \frac{1}{x(n)} + \rho_v h_{fg} \right\}
\]

Notice that the situation can be turned around at this

point in that if the energy deposited by the radiation

for the formation of an embryo is known, the size of the

resulting embryo can be calculated from equation III.5.

This embryo size is one of the required inputs of the

statistical approach of chapter II. The energy of form-
nation of a critical embryo or nucleus is also given by equation III.5 when \( x = 1 \), i.e. \( r_n = r^* \).

\[
E^*(T_1, P_1, \text{Substance}) = \frac{4}{3} \pi r^* \left[ \frac{P^*_b}{2} + \rho_v h_{fg} \right]
\]

Since \( P^*_v = P^*_v - P_1 = 2\sigma/r^* \), the first term on the right hand side of this equation becomes:

\[
\frac{4}{3} \pi r^* \left( \frac{2\sigma}{2r^*} \right) = \frac{4}{3} \pi r^* \frac{2\sigma}{2} = \frac{16\pi \sigma^3}{3(P^*_v - P_1)^2} = W^*
\]

This term is exactly the work of formation of the critical embryo as defined in section II.1. Equation III.6 can also be obtained directly from the First Law of Thermodynamics as shown in Appendix C.

III.4 Available Energy from Radiation

One of the main questions involved in the radiation induced nucleation process which will hopefully be answered by this work is the amount of energy made available from the radiation for the development and growth of any particular embryo. Obviously this will have a considerable effect on the resulting size of the embryo and its ability to become a nucleus as seen in the preceding section.

The interaction of radiation with a substance was
discussed briefly in section II.3. In general the energetic particles involved in this nucleation process can be thought of as producing very long cylindrical regions of energetic molecules along their paths as they interact with the system. It is possible to calculate the quantity of energy deposited in the substance or lost by the particle per unit of distance along the particle track. This quantity is referred to as the energy deposition rate and is a function of the type of radiation, the substance constituting the system and the local energy of the particle.

Since the track length may be several orders of magnitude greater than the critical embryo diameter it is highly unlikely that the total length of the vapor-like cylinder around the track can reconfigure itself into a single sphere of vapor. A much more likely possibility is that this long cylinder in some way becomes fragmented and that these fragments may reconfigure into individual embryos. The situation is shown in figure III.1. The initial track is very small in diameter and probably expands as a cylinder since any break-up during the early stages would tend to expand together again during the diametrical expansion of a couple orders of magnitude. If the final track fragmentation were known, the energy available to a particular embryo could be determined knowing the energy deposition rate.
Figure III.1 Typical Situation for a Heavy Charged Particle Interacting with Matter.

Also shown in figure III.1 is a typical energy de-
position curve for a heavy charged particle interacting with matter. It would appear likely that the greatest energy could be made available to an embryo in the initial part of the track if energy transferred to the electrons remains localized during the energy transfer from the electronic system to molecular motion. If the electrons spread the deposited energy over a large region through electron-electron interactions, it will be essentially unavailable to the embryo. In this case the important portion of the track becomes the nuclear elastic collision portion or the end of the track where the particle's energy is transferred directly into molecular motion.

If a portion of the track of length $\Delta s$ can be assigned to a particular embryo, the energy available to the embryo from the energetic particle is the integral of the energy deposition rate, $\frac{dE}{ds}$, over the length $\Delta s$. In equation III.8 $\Delta E$ is the energy lost by the particle

$$\Delta E = \int_{s_1}^{s_2} \left[ \frac{dE}{ds}(E) \right] ds = E(s_1) - E(s_2)$$

III.8

and as indicated $dE/ds$ is a function of the particle's energy at its position $s$. Since $dE/ds$ is a known function of energy and not position along the track, this integral
cannot be performed directly. An iterative procedure can be employed by calculating the average energy deposition rate over the interval \( E(s_1) - E(s_2) \) and equating it to the energy lost by the particle over the length \( \Delta s \) divided by the length \( \Delta s \). If \( \Delta s \) is known and one of the energy limits is known or assumed, equation III.9 can be

\[
\frac{1}{E(s_2) - E(s_1)} \int \frac{dE}{ds(E)} dE = \left( \frac{dE}{ds} \right) \text{avg. over } \Delta s
\]

solved for the other energy limit and \( \Delta E \) lost by the particle while traversing \( \Delta s \) can be obtained.

The threshold conditions for the occurrence of radiation induced nucleation will be determined by the largest possible value of \( \Delta E \) obtainable along the track. This assumes, of course, that the energy lost by the particle, \( \Delta E \), is equal to the energy available to the embryo, \( E_A \). If it is assumed that the track breaks up in a uniform manner, i.e. \( s = L = \text{constant} \) etc., the maximum value of \( \Delta E \) would occur for \( s = 0 \) to \( s = L \) for the case depicted in figure III.1. With \( E(0) \) known for a particular type of particle and an assumed value of
\( \Delta s = L \), equation III.9 contains only one unknown, \( E(L) \), which can be obtained along with \( \Delta E = E(0) - E(L) \). Particles with different energy deposition characteristics from that shown in figure III.1 would have to be given appropriate special attention.

The actual physical phenomenon involved in the break-up of the vapor wake left by the energetic particle at this point is unknown. Norman (10) suggests the use of the stability condition for vapor jets to determine the break-up of the vapor wake. This condition allows that a vapor jet in a liquid breaks up into discrete regions of length comparable to the circumference of the jet. Then Norman (10) indicates that \( L = 2\pi r_n \), where \( r_n \) is the maximum radius of the embryo under consideration, is an appropriate length to use. In general the minimum value for this length would be \( 2r_n \) since by necessity the embryo will occupy this length of the wake. Rather than to attempt to construct a more detailed basis for the determination of this length at this point it will be put in dimensionless form and carried as a parameter through the theoretical calculations. Then, the comparison of the theoretical results with the data should shed some light on the characteristics of this breakup phenomenon. The parameter defined in equation III.10 should turn out to be approximately constant for all system conditions if the vapor jet condition truly represents the situation.
The maximum available energy for the formation of an embryo is given by equation III.11 subject to equation III.12.

\[ (E_A)_{\text{max.}} = \left[ E(s_2) - E(s_1) \right]_{\text{max. for the track}} - \sum_x \Delta E_x \]  

III.11

\[ E_A = \Delta E - \sum_x E_x = f(T_1, P_1, \text{Radiation, Substance}) \]  

III.12

This assumes that the available energy equals the energy lost by the particle in traversing the path length, \( a r^* \), minus any energy sinks in the track and that this path length, \( a r^* \), is chosen such that \( \Delta E \) is the maximum of the track. Notice that \( s_2 = s_1 + ar^* \) and that \( \Delta E \) is subject to equation III.9.

III.5 Criterion for Radiation Induced Nucleation under the Energy Balance Approach

The criterion for radiation induced nucleation in this approach is very simple and does not depend on the system parameters of volume, radiation reaction rate, and observation interval as was the case for the statis-
tical approach. The criterion is simply that the energy available for embryo formation equal the energy of formation of a critical embryo or nucleus. Substituting equations III.6 and III.11 into equation III.13 subject

\[ E^*(T_1, P_1, \text{Radiation, Substance}) = E_A(T_1, P_1, \text{Radiation, Substance}) \]

III.13

to equations III.9 and III.12 gives the analytical criterion for radiation induced nucleation. The phenomenon of radiation induced nucleation will occur under this

\[ \frac{4}{3} \pi r^3 \left[ \frac{P^* b}{2} + \rho_v h_f g \right] = \left[ E(s_1 + a r^*) - E(s_1) \right]_{\text{max.}} - \sum_x \Delta E_x \]

III.14

approach when the conditions of the system satisfy equation III.14 for a particular value of "a".

III.6 Determination of the Maximum Embryo Size under Non-nucleating Conditions as Prescribed by the Energy Balance Approach

It is desired to determine the maximum size that an embryo can attain for a particular system and system state. This size is a required input of the statistical theory
and in this sense the statistical theory is not a totally independent approach. This can be obtained by again equating the maximum energy available for embryo formation to the required energy of formation of an embryo of size $n$. Then, with equation III.5 equal to III.11 which is subject again to equations III.9 and III.12 the equation for maximum embryo size, $n_R$, from the radiation track is:

\[
\frac{4}{3} \pi r^* 3 x(n_R) \left\{ P_v \left[ 1 - b + b/x(n_R) \right] \ln \left[ 1 - b + b/x(n_R) \right] + \right. \\
\left. \frac{P_v b}{2} \frac{1}{x(n_R)} + \rho_v h_{fg} \right\} = \left[ E(s_1 + ar^*) - E(s_1) \right] - \sum_x \Delta E_x
\]

This equation must be solved by iterative procedures.
Chapter IV

Experimental Program and Experimental Results

In chapters II and III analytical methods were developed for predicting the system conditions for which a particular type of radiation would cause nucleation of the vapor phase of a particular substance. In order to have confidence in these methods it is desirable to apply them to certain cases which can be verified experimentally. Since as a final result of this work it is desired to obtain the conditions under which liquid sodium could be nucleated by radiation, it would, indeed, be helpful to test these analytical methods against a sodium experiment. Due to the complexities involved with sodium experiments in general and, indeed, the questionable feasibility of this particular experiment, the main experimental effort was directed toward water under the influence of fission fragments and fast neutrons.

IV.1 Possible Experimental Approaches

Preliminary calculations for the superheat requirements for water near atmospheric pressure exposed to
fission fragments indicated that superheats in the range of 50 to 100 degrees \( ^{\circ}F \) would be required. Although the superheat requirements would be relaxed at higher pressures, the experimental complexity increases and, additionally, it is desired to test the analytical methods in the low pressure range, 20 to 200 psia. Fission fragments are the best of several types of radiation that could be used from the required superheat standpoint, however, it would certainly be interesting to test the analytical methods against different types of radiation. In general, then, an experimental approach must be used which will provide liquid superheats in this pressure range on the order of 100\(^{\circ}F\).

One approach is a decompression process as utilized in bubble chambers. This method generates superheat as a result of a very rapid drop in liquid pressure at constant temperature. The problem here is the short duration of the highly superheated state. If one is to observe the nucleation in a fission track, the fission rate must be such that a fission event occurs within the duration of the known superheated state. This method has been used successfully by Deitrich (11) for water exposed to fission fragments in the pressure range of 175 to 350 psia. The maximum superheat obtainable in his apparatus was about 20\(^{\circ}F\) and he could not, therefore, investigate the lower pressure range. A photographic
technique was used to determine if the fission events resulted in nucleation for a certain set of conditions. The results of this work will be given along with the results of the present work at the end of this chapter. Deitrich believed that his maximum attainable superheat was limited as a result of competing nucleation phenomenon such as surface defects in the chamber. Then, the only other way in which high superheat may be obtained is to eliminate all heterogeneous nucleation processes such as microscopic surface cavities, suspended matter in the liquid, and very small gas bubbles. With sufficient care the last two nucleation processes can be eliminated, but it is nearly impossible to obtain a solid surface with the required degree of perfection. The answer is to eliminate solid surfaces all together by suspending the liquid under consideration in a second liquid. However, as pointed out by Trefethen (12) the liquid used for suspension purposes must be immiscible in the suspended liquid, must have a density equal to or greater than that of the suspended liquid, and must have a lower volitibility than the suspended liquid. Assuming the proper fluid can be obtained, this method provides the capability of maintaining a highly superheated state for essentially indefinite period of time. This latter method was utilized in this experimental work.
IV.2 Experimental Setup

Trefethen (12) suggests the use of liquid mercury as the suspension fluid. However, problems with the handling of this material and problems of maintaining a clean surface for repeated experiments motivated a search for some other material. It turned out that a silicone oil under the trade name of "Dow Corning 550 Fluid" was very satisfactory. Feasibility experiments showed that high superheats greater than 100°F could be generated at atmospheric pressure, that fission fragments did, indeed, nucleate the superheated liquid, and that macroscopic boiling could be visually observed. This preliminary data contained considerable scatter which was believed to be caused by the inaccuracy in the temperature measurements. Due to the fact that the drop of water was not totally submerged in the suspension fluid it was continuously evaporating thus making it somewhat cooler than the surrounding oil. Since the temperature of the water cannot be measured directly, attempts to deduce the water temperature from the surrounding oil temperature were difficult and inaccurate. Also due to the continuous evaporation the drop had a relatively short lifetime before it was considerably diminished in size which resulted in other experimental difficulties. The evaporation problem was easily corrected by adding a
cover fluid above the suspension oil and the drop. It was found that heavy weight mineral oil was satisfactory for this purpose. It was also found, however, that temperature gradients still persisted in the oil surrounding the drop and that the actual drop temperature would have to be deduced from the oil temperature in some appropriate manner.

The experimental setup is shown in figures IV.1 and IV.2. The system is designed so that the nucleation phenomenon can be investigated at various pressures from atmospheric up to about 100 psia. The setup includes the boiling chamber, various injection and ejection capabilities, pressure gauge, thermocouple with associated potentiometer and recorder, convection current generator for maintaining the water drop in the center of the chamber and directly over the thermocouple, electric heater, and cooling system. The injection and ejection capabilities are required for inserting the water drop into its suspended position, for adjusting the drop's vertical position by varying the silicone oil level, for applying the cover oil layer in the chamber, for removing moisture from the chamber, for removing the remaining drop or drops after an experiment, and removing cover oil from the chamber after the experiment.

The experimental setup can be best explained in detail by describing the experimental procedure for a
Figure IV.1 Experimental Setup for Determining the Superheat Threshold for Water Exposed to Radiation.
Figure IV.2 Boiling Chamber.
single run. It can be assumed that the water flow rates
through the vapor condenser and convection generator are
set at their appropriate values, the two oil reservoirs
and the water reservoir are filled, the pressure in the
boiling chamber is atmospheric, valves 1 through 6 and
also 9 and 10 are closed, and the silicone oil in the
chamber is at a temperature less than 212°F. With the
system in this condition, valve 4 is opened, the silicone
oil reservoir is pressurized by valve 10, valve 6 is
opened, the silicone oil level in the boiling chamber is
brought up to the insertion tubes, and valve 6 is closed.
Then the cover oil is added by opening valves 1 and 2
allowing the oil to flow until a layer approximately \( \frac{1}{4} \)
inch thick is formed in the boiling chamber. These two
valves are then closed and the water drop is inserted.
This is accomplished by very carefully opening valve 3
and allowing the water to very slowly flow down to the
interface between the two oils. If the flow is too fast,
the water does not have time to acquire the same tempera-
ture as the oil and the colder water sinks in the oil
as it flows in without forming a single large drop.
When the drop becomes about \( \frac{1}{8} \) inch in diameter, valve 3 is
closed and the oil level is lowered to remove the drop
from the insertion probe and to place it as near as
possible to the thermocouple probe but not contacting it.
Valve 4 is then closed and pressurization through valve 9
takes place, if the run is a pressure run, followed by heating at any desired rate to the saturation temperature. During all heating periods the atmosphere in the boiling chamber becomes hotter and, therefore, the pressure increases in the system if the system is completely tight. However, a constant pressure system can be maintained by setting the air supply pressure valve, 10, to the desired pressure and allowing the system to leak very slightly. The supply valve will then make up the leak rate and maintain the system at the desired pressure. Also as heating takes place the suspension oil expands tending to raise the water drop away from the thermocouple probe. It is necessary, then, to frequently adjust the oil level by opening valve 6 and allowing oil to flow by gravity out of the chamber until the drop is lowered to its appropriate position. With the system at the saturation temperature for the particular pressure of interest and with the drop in position the experiment is ready to begin.

Continuation of the experimental run from the saturation conditions involves adjusting the heating rate to provide the desired temperature ramp, adjusting the suspension oil level, observing the drop inside the boiling chamber through the glass window, and making any necessary adjustment in the convection generator flow rate to maintain the drop in a central position in the
The convection generator is simply a small cooling coil in the center of the boiling chamber. The oil which is cooled by this coil flows downward in the center of the chamber and upward along the outer heated walls of the chamber. Then at the top of the oil pool the oil flow is radially toward the center which keeps the drop from drifting out toward the walls of the chamber and at the same time supplies heat to the drop. Since the water flowing in this coil is at atmospheric pressure, it boils at the operating conditions of the chamber causing flow changes and necessitating periodic monitoring.

At low pressure the nucleation phenomenon is easily observable since the drop literally explodes due to the large amount of stored energy in the superheated liquid and the large specific volume change associated with the phase change. It might be noted that the limit on pressure which may be investigated using this system is the pressure for which the macroscopic bubbles resulting from the nucleation process are so small that they are unobservable through the windows of the chamber. When the boiling event occurs the temperature is recorded and the system is cooled down to begin the cleanup process.

Since the drop injection procedure described above must take place at atmospheric pressure, the system must be cooled down and depressurized. The cooling is done by
allowing full flow through the convection generator after shutting off the heater. The pressure is released through valve 4 after closing valve 9. Note that when the pressure is released through valve 4 any condensate in the condensate pail within the chamber is ejected. The oil level is again raised as indicated above using valve 6 until the interface of the two oils is approximately at the ejection probe. Valve 4 is closed and the boiling chamber is slightly pressurized. Then valve 5 is opened allowing the exploded drop and cover oil to be forced out of the chamber. It may be necessary at this point to remove moisture from the silicone oil and portions of the exploded drop which may have come into contact with the solid surfaces in the chamber. To do this the system is simply heated up at atmospheric pressure to drive off the moisture which is then condensed by the condensing tube at the top of the chamber with the condensate dropping into the small pail. The system is then cooled down and made ready for another run.

IV.3 Pressure Gage and Thermocouple Calibration

The system pressure in this experimental setup was measured by a 0 to 100 psig Bourdon gage. Since an accurate value of the pressure was necessary particular-
ily at low pressure in order to determine the saturation temperature and, consequently, the superheat from the saturation temperature and the measured liquid temperature, it was necessary to accurately calibrate the pressure gage. This was done using a Refinery Supply Company "Dead Weight Tester". The calibration results are shown in table IV.1.

<table>
<thead>
<tr>
<th>Dead Weight Tester (psig)</th>
<th>Pressure Gage (psig)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>12.5</td>
</tr>
<tr>
<td>20</td>
<td>22.0</td>
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<td>30</td>
<td>31.5</td>
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<td>41.0</td>
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<td>70</td>
<td>70.5</td>
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<tr>
<td>80</td>
<td>80.5</td>
</tr>
<tr>
<td>90</td>
<td>91.0</td>
</tr>
<tr>
<td>100</td>
<td>101.0</td>
</tr>
</tbody>
</table>

The oil temperature directly below the drop was measured with a Chromel-Alumel thermocouple placed inside an aluminum sheath. The thermocouple potential was measured both by a Leed and Northrup Type K-3 Universal Potentiometer and a Minneapolis-Honeywell strip chart recorder with a chart range of 0 to 1 millivolts. The recorder was capable of being biased in 1 Mv steps.
so that any potential could be recorded with the same scale. The recorder was the actual recording instrument during the experiments with the potentiometer serving as a check. In this way a specific temperature ramp could be established by adjusting the heater power to obtain a particular slope of the temperature trace.

The temperature measuring system was calibrated as a system by measuring the boiling points of water and aniline at atmospheric pressure. The system measured these temperatures as 212.0°F and 363°F, respectively. The agreement is very good for water and the measured value for aniline compares well with the value of 364.1°F given by Perry (13). It appears that the temperature measuring system is sufficiently accurate for the experiments to be done in this work.

IV.4 Temperature Correction Due to Temperature Gradients in the Oil

In the preliminary experiments to test the feasibility of this system it was found that the temperature being recorded by the thermocouple was very sensitive to the distance between the bottom of the drop and the thermocouple probe even under steady state conditions. This effect was believed to be due to the evaporation of the drop as discussed above. The addition of the cover oil
appeared to remedy this problem and the probe gave nearly the same temperature readings for distances of 1/16 inch to almost touching the drop, whereas, previously the readings changed several degrees over this range of distances. This led to the assumption that the thermocouple probe was reading essentially the temperature of the drop.

Near the end of the experimental program a piece of dirt was noticed inside a drop, and it could be seen following a natural convection type path, i.e. upward along the outside edge of the drop and downward in the center of the drop. This observed flow indicated that a temperature gradient existed in the oil surrounding the drop with the colder temperature at the top of the drop. The natural convection currents also indicated that temperature variations existed in the drop and that the average temperature of the drop would probably be somewhat less than the temperature being measured by the thermocouple probe.

It was decided to measure the temperature distribution in the oil around the drop to see what the situation was. The cover oil insertion probe was removed and a vertically translating thermocouple probe was inserted in its place. By rotating the probe while at any vertical location the thermocouple junction could be located at the surface of the drop as shown in figure IV.3. Several temperature traverses were made at five different vertical
locations from the top of the drop to the bottom of the
drop which was the location of the stationary thermo-
couple probe. The measurements were by necessity made
atmospheric pressure and had to be made without a drop

Vertically Translating and Rotating
Thermocouple Probe

Thermocouple Junction

Cover Oil

Convection Currents

Suspension Oil

Stationary Thermocouple Probe

Figure IV.3 Thermocouple Arrangement for Measuring Temperature Distribution Around the Water Drop.

present in the higher temperature range, 375°F, since
for atmospheric pressure and this temperature 163°F super-
heat would be required, which would be very difficult to
achieve. Even in the low temperature range, 250°F, care
had to be taken not to disturb the drop with the probe or
it would boil. Temperature measurements in the low
temperature range without a drop present did not show
appreciable difference from these taken with the drop
present.

Figure IV.4 shows the resulting temperature distribu-
tion around the drop. The distribution for the tempera-
ture range of 250°F and that for the temperature range of
375°F, which fairly well bracket the temperatures of
interest in this work, are very nearly the same except
in the interface region, \( x \approx \frac{1}{2} \) inch. The difference
could very well result from the fact that a drop was not
present in the high temperature case. If a drop were
present it would in effect pump heat from the hot region
at the bottom of the drop to the interface region at the
top. This would tend to decrease the oil temperature
around the bottom of the drop and increase the oil temp-
erature near the top, thus bringing the high temperauure
curve more in agreement with the low temperature curve.
The temperature seems to drop off rapidly near the inter-
face region between the two oils. It appeared that the
convection flow in the suspension oil which maintains the
drop location and transfers heat to the drop was not
doing a very effective job of heating the cover oil. The
interface between the oils appeared to be acting as a
separating surface and the heat transfer to the cover oil
was by conduction only. Because of the relatively low
Figure IV.4 Measured Temperature Distribution Around the Drop Surface.
thermal conductivity of the stagnant cover oil it was acting as an insulator between the hot suspension oil and the atmosphere in the upper boiling chamber, thereby setting up temperature gradients of considerable magnitude. Had this problem been recognized at the beginning of the experimental program, some sort of heating system could have been put in or around the upper part of the boiling chamber to more nearly equalize the temperature throughout the system.

From the temperature distribution around the drop an attempt will be made to establish a realistic correction factor to apply to the experimental temperature measurements. If the spherical drop is considered as a small heat pump which receives energy from the hot oil around the lower portion of the drop and transfers this energy to the cold oil around the upper portion of the drop, then, at steady state the heat transferred to the drop must just equal the heat transferred from it. Assume that the water in the drop is sufficiently well mixed that it can be described by a mean temperature throughout, $T_m$. Also assume that the heat transfer coefficient, $h$, for heat transfer from the drop surface to the water is uniform over the drop. Then the heat transferred through an element of surface, $dA = 2\pi r_d dx$, as shown in figure IV.5 is:
\[ dq = h(2\pi r_d dx) \left[ T_s(x) - T_m \right] \]  \hspace{1cm} \text{IV.1}

The total heat input to the drop can be found by integrating equation IV.1 from \( x = 0 \) where \( T_s(0) = T_p \) to \( x = x_R \) where \( T_s(x_R) = T_m \). \( x_R \) is the vertical location where the heat transfer is reversed, hence, the subscript \( \text{TNNatural Convection} \)

\[ \text{Currents} \]

\[ \text{Suspension Oil} \]

\[ \text{Stationary Thermocouple Probe} \]

\[ \text{Figure IV.5 Model for Calculating the Actual Mean Temperature of the Drop.} \]

R. Similarly the total heat rejected by the drop can be found by integrating equation IV.1 from \( x = x_R \) where \( T_s(x_R) = T_m \) to \( x = \frac{1}{2} \) where \( T_s(\frac{1}{2}) = T_T \).
Then using the measured surface temperature distribution for $T_s(x)$ one can find the $T_m$ which satisfies equation IV.3. This can be done graphically using figure IV.4. The uniform mean drop temperature is a horizontal line on this figure. The correct value of the mean temperature is the one which makes area $A_1$ equal to area $A_2$ as prescribed by equation IV.3. For the measured distributions the mean drop temperature was found to be approximately $7.5^\circ\text{F}$ below the temperature measured by the probe, $T_p$. Therefore, with the apparent constancy of the surface temperature distribution with system temperature level, a constant correction factor of $-7.5^\circ\text{F}$ will be applied to all the experimental temperature data.
IV.5 Determination of Fission Rate and Temperature Ramp for the Fission Fragment Experiments

The first series of experiments were designed to find the superheat as a function of pressure at which fission fragments would nucleate the phase change in water. The fission fragments were obtained by irradiating a dilute solution of uranium nitrate with neutrons from five 1 Ci PuBe neutron sources arranged outside the boiling chamber. This setup allowed the fission events to take place directly in the superheated solution, thereby permitting the total energy of the fragments to participate in the process.

A valid question arises at this point as to the effect of the solution on the process being investigated. One would expect that the solute would primarily effect the surface tension, and the magnitude of this effect on the required superheat for nucleation will be discussed in chapter VII. In order to minimize the deviation of this experiment from the pure water case, the solution should be made as dilute as possible. The concentration of $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ in the solution also directly affects the fission rate in the drop and the operating procedure for the experiment. Therefore, its concentration should be selected based on both considerations which turn out to be somewhat conflicting.

The fission rate in the drop is given approximately
by equation IV.4 where $N_u$ is the uranium atom density, 
$ar{\sigma}$ is an average cross-section for fission, $\phi$ is the total neutron flux at the drop, and $V_d$ is the volume of the drop.

$$F.R. = N_u \bar{\sigma} \phi V_d \quad \text{IV.4}$$

It is not important to know this fission rate to a high accuracy so some simplifying assumptions can be made.

Although the neutron sources produce neutrons with a spectrum of energies up to about 10.5 Mev as indicated by Stewart (14) and the cross-section is energy dependent, a reasonable estimate for the fission rate can be obtained by taking the value of the fission cross-section for U$^{238}$, since natural uranium is being used, at approximately 4 Mev. Lontai (15) indicates that the fission cross-section for U$^{238}$ in the neutron energy range from about 2 Mev to 6 Mev is nearly constant and equal to about 0.55 barns ($10^{-24}$ cm$^2$). Then, using this value of $\bar{\sigma}$ with the total integrated flux should give a reasonable approximation of the fission rate.

The total flux at the drop is equal to the total number of neutrons emitted per second by the five 1 Cu PuBe sources divided by a spherical surface area of radius equal to the distance between the sources and the drop. This distance in this experimental setup is about 11 cm and the total source strength of approximate-
ly $8 \times 10^6$ neutrons/sec from Karaian (16). Assuming negligible attenuation of the flux due to the structural material and oil between the sources and the drop, the total flux at the drop is approximately $0.53 \times 10^4$ neutrons per cm$^2$ per sec.

The atom density of $^{238}U$ in the solution used for these experiments is given by equation IV.5 where $\rho_U$ is the density of $^{238}U$ in grams/cm$^3$, $N_A$ is Avagadro's number and $M_U$ is the molecular weight of $^{238}U$. The density of $^{238}U$ is proportional to the concentration

$$N_U = \frac{\rho_U N_A}{M_U} \quad \text{IV.5}$$

of solute in the solution and is given by equation IV.6 where $C_S$ is the mass of solute per unit mass of water.

$$\rho_U = \frac{C_S M_U}{M_S} = 0.474 C_S \quad \text{IV.6}$$

With the diameter of the drop taken as 0.5 inches the fission rate as a function of solute concentration becomes:

$$F.R. = \frac{0.474 C_S}{238} 0.6023(10^{-24}) 0.53(10^4) \frac{1}{6} \pi [0.5)(2.54)] 30.55(10^{-24})$$

$$= 3.77 C_S \text{fission/sec} = 227 C_S \text{fission/min.} \quad \text{IV.7}$$
The fission fragments resulting from these fissions will not be the same for each event but will constitute a whole spectrum of elements with mass number ranging from 70 to 160. The yields of these various fission fragments which actually appear as pairs are highest for pairs having mass numbers of approximately 97 for the light fragment and 142 for the corresponding heavy fragments as indicated by Evans (17). Since the majority of all the resulting fragments pairs are in this range of mass number, 97±5 and 142±5, and since the fragments in this narrow band can be considered essentially identical from the energy deposition standpoint, it seems reasonable to assume that most of the fission events will result in fragments that can be considered characteristic of the most probable, i.e. 97 and 140.

The procedure for actually performing the experiment once the apparatus is in its appropriate condition as indicated in section IV.2 is to put the system on a specified temperature ramp and observe the temperature at which boiling occurs. Then, depending on the accuracy desired for the threshold temperature, the temperature ramp, TR, should be specified for a particular fission rate such that at least one fission event occurs in the time interval for the temperature to rise by the amount of the maximum desired uncertainty, $\delta T^0_F$. 
(F.R.) \( \delta T \geq TR \)

It would seem unrealistic to expect to determine the superheat threshold to an accuracy of less than \( 1^\circ F \). Then, with \( \delta T = 1 \), any ratio of temperature ramp, \( TR \), to fission rate equal to unity should give satisfactory results. For a temperature ramp of \( 1^\circ F/\text{min} \) which was easy to maintain experimentally, a fission rate of at least 1 per min. is desirable. To establish this fission rate a solute concentration of less than 1\% by mass is required which will be shown in chapter VII to be acceptable from a surface tension point of view. The actual concentration used throughout the fission fragment experiments was 0.0087 gm solute per gm water.

IV.6 Experimental Data for Fission Fragments in Water

Data was taken at approximately 10 psi intervals from 14.7 psia to 94.2 psia. At each pressure a number of identical experimental runs were made to indicate the reproducability of the results. Additional runs were made between data runs to insure that the boiling events were indeed fission fragment induced and not spurious. These latter runs were done by following the same procedure as for a data run except the neutron sources were not used and, therefore, the fission fragments were not generated. In
nearly all cases no boiling was observed for temperatures on the order of 15 to 20°F above the apparent threshold. In most of these cases the run was simply terminated in order to avoid the cleanup problems that resulted when boiling occurred at high superheat. Occasionally a run would have to be terminated because persistent boiling occurred on the way up to the apparent threshold. These were taken to be drops containing some type of dirt which often could be confirmed visually.

In table IV.2 are listed the data obtained for fission fragments in water. A mean superheat heat is listed for each pressure along with the number of runs on which the mean is based. Also given is the standard deviation of the data taken at each pressure. As can be seen the threshold appears to be very well defined and reproducible. These standard deviations are well within the probably overly optimistic overall experimental accuracy of 1°F. The last column of the table gives the mean superheats after applying the -7.5°F correction factor.

Deitrich (11) obtains the superheat threshold for fission fragments in water with his bubble chamber work for pressures somewhat higher than the ones in table IV.2. His data for the minimum superheat observed at the particular pressure is listed in table IV.3. Both these sets of data are plotted in figure IV.6. The general trends in
Table IV.2  Experimental Superheat Data for Fission Fragments in Water.

<table>
<thead>
<tr>
<th>Pressure (psia)</th>
<th>Number of Runs</th>
<th>$T_{sat}$ (°F)</th>
<th>$\Delta T_{mean}$ (°F)</th>
<th>$\sigma$ (°F)</th>
<th>$(\Delta T_{mean})_{corr}$ (°F)</th>
</tr>
</thead>
<tbody>
<tr>
<td>14.7</td>
<td>10</td>
<td>212.4</td>
<td>62.6</td>
<td>0.3</td>
<td>55.1</td>
</tr>
<tr>
<td>24.7</td>
<td>10</td>
<td>239.4</td>
<td>53.1</td>
<td>0.6</td>
<td>45.6</td>
</tr>
<tr>
<td>32.7</td>
<td>10</td>
<td>255.8</td>
<td>46.5</td>
<td>0.5</td>
<td>39.0</td>
</tr>
<tr>
<td>43.2</td>
<td>14</td>
<td>271.9</td>
<td>40.8</td>
<td>0.7</td>
<td>33.3</td>
</tr>
<tr>
<td>53.7</td>
<td>10</td>
<td>285.5</td>
<td>37.0</td>
<td>0.7</td>
<td>29.5</td>
</tr>
<tr>
<td>63.7</td>
<td>12</td>
<td>296.6</td>
<td>36.2</td>
<td>0.6</td>
<td>28.7</td>
</tr>
<tr>
<td>74.2</td>
<td>10</td>
<td>306.9</td>
<td>32.1</td>
<td>0.6</td>
<td>24.6</td>
</tr>
<tr>
<td>84.2</td>
<td>11</td>
<td>315.6</td>
<td>28.5</td>
<td>0.5</td>
<td>21.0</td>
</tr>
<tr>
<td>94.2</td>
<td>10</td>
<td>323.5</td>
<td>28.5</td>
<td>0.6</td>
<td>21.0</td>
</tr>
</tbody>
</table>

The data and the comparison with theoretical predictions will be discussed in another chapter.

Table IV.3  Deitrich's Data for Minimum Superheat at the Given Pressure.

<table>
<thead>
<tr>
<th>Pressure (psia)</th>
<th>$\Delta T$ (°F)</th>
<th>Uncertainty $\pm$ (°F)</th>
</tr>
</thead>
<tbody>
<tr>
<td>176</td>
<td>12.4</td>
<td>0.64</td>
</tr>
<tr>
<td>190</td>
<td>12.0</td>
<td>0.61</td>
</tr>
<tr>
<td>210</td>
<td>10.7</td>
<td>0.57</td>
</tr>
<tr>
<td>248</td>
<td>10.3</td>
<td>0.46</td>
</tr>
<tr>
<td>288</td>
<td>10.4</td>
<td>0.41</td>
</tr>
<tr>
<td>346</td>
<td>10.2</td>
<td>0.37</td>
</tr>
</tbody>
</table>
Figure IV.6 Experimental Superheat Data from this Work and from Deitrich's Work.
IV.7 Experimental Procedure for Fast Neutrons in Water

Experiments using fast neutrons without the production of fission fragments were performed using pure water instead of the uranium solution. The nucleation phenomenon in this case is brought about by the primary knock-on atoms produced in elastic scattering reactions between the energetic neutrons and the atoms of the system. In the case of water the primary knock-ons would be the hydrogen and oxygen atoms. The relative importance of these two secondary energetic particles for nucleation purposes will be discussed in more detail in chapter VI. It will suffice for the moment to indicate that the oxygen knock-ons will be of primary importance.

From an experimental standpoint the only difference between fast neutrons with associated PKOA's, primary knock-on oxygen atoms, and the fission fragments is in the amount of superheat required at the same system pressure. However, in analyzing the results for theoretical comparison much more difficulty is encountered. This difficulty is a result of the polyenergetic characteristics of the PKOA's and the sensitivity of their energy deposition rates to their energy. Qualitatively this comes about due to neutron energy spectrum of the fast neutrons from the PuBe neutron sources and due to possibility in any particular scattering event for the neutron to be
scattered into any angle from 0 to 180°. This latter
effect means that the neutron may transfer to the PKOA
a maximum amount of energy in the case of a "head-on
collision" or it may transfer very little in a small
angle scattering. Then, since there are very few neutrons
from the sources with maximum energy and only a small
fraction of these undergo "head-on collisions" with
oxygen atoms, there will be a very small number of PKOA's
produced with the maximum possible energy. These are the
PKOA's, however, which determine the absolute threshold
conditions for fast neutron induced nucleation. If one
desired to measure these absolute threshold conditions in
an experiment, a very intense neutron source or a very
long duration experimental run would be required. In
the experimental apparatus used in this work measurement
of this absolute threshold was not feasible due to the
relatively low intensity neutron source and the difficulty
of maintaining an essentially zero temperature ramp for
very long periods of time, on the order of hours.

The effect of a realistic temperature ramp is to
by-pass the range of superheat where the production rate
of eligible PKOA's is low, thereby proceeding to higher
superheats. This is, of course, near the absolute thresh-
hold condition. By eligible PKOA's is meant those with
sufficient energy to induce nucleation at the particular
system state. The net result of a reasonable temperature
ramp is to shift the data toward higher superheat and in effect make lower energy PKOA's eligible for the process. Then, for any set of experimental data taken at some particular temperature ramp the actual effective PKOA energy participating in the process is unknown and analytical comparisons cannot be made. Therefore, the experimental program for fast neutrons in water was expanded from just taking data at various pressures and a particular temperature ramp to also taking data at various temperature ramps at a particular pressure. Using this information in the analysis of chapter VI will allow a determination of the effective participating PKOA energy for the conditions under which the data at various pressures was taken. With this effective energy theoretical superheats as a function of pressure can be generated and realistic comparisons made in chapter VII.

It should be pointed out that this ramp rate effect is pronounced only at very low reaction rates such as encountered in this particular experimental setup. In the fission fragment case the light fragments could be assumed identical, thereby providing a much higher number of eligible particles to work with.

The actual experimental procedure used in this set of experiments was exactly the same as for the fission fragments. Various runs were made without neutrons present to check the authenticity of the events observed
and, as before, these runs nearly always attained superheats considerably above the apparent threshold before boiling occurred or the run was terminated. A number of runs were made at each pressure from 55 psia to 95 psia at 20 psi intervals at approximately the same temperature ramp. Three ramps of approximately $\frac{1}{2}$, 1, and 2 degree F per minute were investigated at a pressure of 75 psia.

IV.8 Experimental Data for Fast Neutrons in Water

Preliminary experimentation indicated that sufficiently high superheats could not be consistently obtained for fast neutron induced nucleation at pressures below 50 psia. Therefore, data was taken at a temperature ramp of approximately $\frac{1}{2}$ degree F per minute for three different pressures. This data is presented in table IV.4 in terms of the mean superheat at which boiling occurred and the mean temperature ramp over the number of runs listed. Also listed are the standard deviations in the superheat and temperature ramp and the corrected mean superheat. The data at various temperature ramps was taken at 75 psia. The results are shown in table IV.5 along with actual temperature ramps and standard deviations.
Table IV.4 Experiment Data for Fast Neutrons in Water at Various Pressures.

<table>
<thead>
<tr>
<th>Pressure (psia)</th>
<th>Number of Runs</th>
<th>( \Delta T_{SUP} ) m</th>
<th>( T_R ) m</th>
<th>( \sigma_{T_{SUP}} )</th>
<th>( \sigma_{T.R.} ) Corrected</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>( ^\circ F )</td>
<td>( ^\circ F )</td>
<td>( ^\circ F )</td>
<td></td>
</tr>
<tr>
<td>55</td>
<td>7</td>
<td>114.0</td>
<td>0.54</td>
<td>0.5</td>
<td>0.08</td>
</tr>
<tr>
<td>75</td>
<td>8</td>
<td>96.9</td>
<td>0.36</td>
<td>0.7</td>
<td>0.07</td>
</tr>
<tr>
<td>95</td>
<td>7</td>
<td>91.9</td>
<td>0.56</td>
<td>1.5</td>
<td>0.09</td>
</tr>
</tbody>
</table>

Table IV.5 Experimental Data for Fast Neutrons in Water for Various Temperature Ramps.

<table>
<thead>
<tr>
<th>Pressure (psia)</th>
<th>Number of Runs</th>
<th>( \Delta T_{SUP} ) m</th>
<th>( T_R ) m</th>
<th>( \sigma_{T_{SUP}} )</th>
<th>( \sigma_{T.R.} ) Corrected</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>( ^\circ F )</td>
<td>( ^\circ F )</td>
<td>( ^\circ F )</td>
<td></td>
</tr>
<tr>
<td>75</td>
<td>8</td>
<td>96.9</td>
<td>0.36</td>
<td>0.7</td>
<td>0.07</td>
</tr>
<tr>
<td>75</td>
<td>8</td>
<td>102.5</td>
<td>1.19</td>
<td>0.6</td>
<td>0.05</td>
</tr>
<tr>
<td>75</td>
<td>11</td>
<td>107.7</td>
<td>2.0</td>
<td>1.5</td>
<td>0.13</td>
</tr>
</tbody>
</table>

These two sets of data will be dealt with more fully in chapters VI and VII where the effects of temperature ramp will be considered and comparisons with theory will be made.
CHAPTER V

Application of Statistical and Energy Balance Methods to the Case of Fission Fragments in Water

In this chapter the two theoretical approaches to the phenomenon of radiation induced nucleation will be applied to a particular case. This case will be fission fragments in water for the conditions experienced in the experiments described in chapter IV. This will define the system variables of volume, radiation reaction rate, observation time, substance, and type of radiation. The system pressure and the dimensionless track length "a" will be carried as parameters leaving only the liquid temperature or superheat as the unknown.

The energy balance method involves the solution of equation III.14 subject to equations III.9 and III.12. The statistical method involves the solution of equation II.72 with equations II.61, II.67, and III.15.

V.1 Energy Deposition by Fission Fragments

Both theoretical methods employed here require a knowledge of the energy deposition rate of the fission frag-
ment as it passes through water. Knowing this relationship as a function of fragment energy permits the use of equation III.9 to find the energy deposited in a certain region, $\Delta E$.

The nature of the fission fragment is such that the classical theory of heavy charged particle interaction with a substance is considered reasonably good. Segre (18) gives the expression shown in equation V.1 for the energy deposition rate or the energy deposited in the substance per unit length along the path of the energetic particle. Since water is made up of two types of atoms, the energy deposition to both hydrogen and oxygen is considered separately and added together.

$$\frac{1}{N} \frac{dE}{ds} = \frac{4\pi e^4}{m_0 V^2} (Z_1)^2 \sum_{i=0\&H} \nu_i Z_1 \ln \frac{1.123m_0 V^3}{(I_1/\hbar)e^2(Z_1)^2}$$

$$+ \frac{4\pi e^4 (Z_1)^2}{V^2} \sum_{i=0\&H} \nu_i \frac{Z_1^2}{M_1} \ln \frac{M_1 M_1 V^2 (a_{scf})}{(M_1+M_1)Z_1 Z_1 e^2}$$

V.1

The symbols have the following meanings:

- $e$ = charge on an electron
- $N$ = number of molecules of stopping medium per unit volume
\( m_0 \) = electron mass

\( V \) = velocity of the fission fragment

\((Z_1)_{\text{eff}} \) = effective charge on the fragment = \( f(v) \)

\( Z_1 \) = atomic number of the fragment

\( \nu_i \) = number of \( i \)th atoms per molecules.

\( I_i \) = mean ionization potential \((I_H = 15.5, I_0 = 100)\)
of the \( i \)th component from Evans (17)

\( M_1 \) = mass of the fragment

\( M_i \) = mass of the \( i \)th atom in amu.

\( a_{i1}^{scr} \) = impact parameter beyond which energy loss by the
fragment is effectively zero due to screening of nuclei by atomic electrons for the \( i \)th atom.

\( \hbar = \hbar/2\pi \) = Planck's constant divided by \( 2\pi \)

The effective charge on the fission fragment changes as the velocity of the fragment decreases due to the capture of electrons from the stopping medium. An approximate relationship for this effect from Segre (18) is:

\[
(Z_1)_{\text{eff}} = (Z_1)^{1/3} \frac{\hbar V}{e^2}
\]

The impact parameter \( a_{i1}^{scr} \) can be obtained using the following expression from Claxton (19),
\[ a_{\text{scr}}^\text{li} = \frac{a_H}{\left( (z_1^{2/3} + (z_1^{2/3})^2 \right)^{1/2}} \quad \text{(V.3)} \]

where \( a_H \) is the radius of the first Bohr orbit for the hydrogen atom. The molecule density in the stopping medium is given by the following expression.

\[ N = \frac{\rho_1(T_\text{l}) N_Av}{M} \quad \text{(V.4)} \]

where \( N_Av \) is Avagadro's number, \( \rho_1 \) is the liquid density and \( M \) is the molecular weight of the stopping medium.

There is a very large number of possible fission fragments pairs which can result from fission as discussed in section IV.5 but the most probable pair will be the only one considered. This particular pair, generally referred to as the light and heavy fragment, has the following characteristics as given in Evans (17).

Light fragment: \( A = 97 = M_1^L \)

\[ E_{\text{initial}} = 95 \text{ Mev} \]

\[ Z = 38 = Z_1^L \]

\[ V_{\text{initial}} = 1.4 \times 10^9 \text{ cm/sec} \]
Heavy fragment: \[ A = 138 = N_1^H \]
\[ E_{\text{initial}} = 67 \text{ Mev} \]
\[ Z = 54 = Z_1^H \]
\[ V = 0.93(10^9) \text{ cm/sec} \]

Here the superscript \( L \) stands for the light fragment and \( H \) for the heavy.

Combining equations V.2, V.3, and V.4 with V.1, substituting the appropriate values for the quantities involved, and using the relation \( E = \frac{1}{2} M_1 V^2 \), the following equations for the energy deposition rates of the fragments are obtained.

\[
\frac{dE}{ds}^L = \rho_1(T_1) \left[ 3.620 \times 10^4 \ln(0.0549E) + 3.666 \times 10^3 \ln(8.096E) \right]
\]

\[ V.5 \]

\[
\frac{dE}{ds}^H = \rho_1(T_1) \left[ 4.577 \times 10^4 \ln(0.0343E) + 10.51 \times 10^3 \ln(3.697E) \right]
\]

\[ V.6 \]

The first terms in these two equations represent the energy deposition to the electronic system of the stopping medium through ionization and excitation of the atoms. The second terms represent the energy deposition to the medium.
through nuclear elastic collisions between the fragments and the nuclei of the stopping medium. There is some question in the case of energy deposition to the electronic system of a substance as to the fate of the deposited energy. It is possible for it to be transferred to molecular motion in which case it appears as thermal energy or it can be transferred to other electrons in the system through electron-electron interactions and effectively spread throughout the system. This latter possibility would not result in the formation of a track of highly energetic molecules in the thermal sense. The predominance of the electron-electron mechanism would be expected in materials such as metals where the electrons are primary transporters of energy. This possibility will have to be considered when the theory is applied to liquid metals. In the case of water it will be assumed that the energy transferred from the fragments to the electronic system of the water will result in a track of energetic molecules, thus satisfying equation III.12. Then looking at equations V.5 and V.6 one can see that the first terms predominate at high energy and that they go to zero at 18.2 Mev and 29.1 Mev respectively. Then for energies below these, the second terms only need be considered. Since the highest average value of dE/ds is desired and dE/ds increases with energy only the high energy portion of the track, where
energy deposition by nuclear elastic collisions can be neglected, will be considered.

\[
\begin{align*}
\frac{dE}{ds}^L & = \rho^L(T_1) 3.620(10^4)\ln(0.0549E) \text{ MeV/cm} \\
\frac{dE}{ds}^H & = \rho^H(T_1) 4.577(10^4)\ln(0.0343E) \text{ MeV/cm}
\end{align*}
\]

V.7

V.8

The energy deposition characteristics of the fission fragments are seen from equations V.5 and V.6 to be of the type shown in figure III.1. Then in order to maximize the energy deposited in a length of track, ar*, the portion of the track from \( s = 0 \) to \( s = ar* \) should be considered. Comparing the energy deposition rate of the light and heavy fragments indicates that the light fragment has a higher \( dE/ds \) and will give a higher \( \Delta E \). Since it is desired to maximize \( \Delta E \) for a length, \( ar* \), only the light fragment need to considered in determining the threshold conditions of radiation induced nucleation. Considering equation III.9 the following is known:

\[
E(s_1) = E(0) = 95 \text{ MeV}
\]

V.9

\[
\frac{dE}{ds}(E) = \text{equation V.7}
\]
Equation III.9 can be solved for $E(s_2)$ for any prescribed liquid pressure, liquid temperature, and parameter "a".

\[
\frac{1}{E(s_2) - 95} \int_{95}^{E(s_2)} \rho_1(T_1) 3.620(10^4) \ln(0.0549E) dE = \frac{95 - E(s_2)}{a^*}
\]  

or

\[
a^*(T_1, P_1) \rho_1(T_1) 10^4 = \frac{[95-E(s_2)]^2}{172.6+14.13E(s_2)-3.620E(s_2)lnE(s_2)}
\]

The trial and error solution of this equation then gives the energy deposition for the nucleation process.

\[
\Delta E = 95 - E(s_2) \text{ MeV}
\]

V.2 Effects of the Radiolysis of Water

The generation of hydrogen gas in the fission track by radiolysis of water affects the superheat threshold theory...
in two ways. First, the reaction which produces the \( \text{H}_2 \) gas is an endothermic reaction and therefore, acts as an energy sink in the fission track, tending to reduce the available energy for nucleation. This problem is taken care of in the general theory by the \( \Delta E_\text{x} \) term in equation III.14. The other effect is to change the dynamic equilibrium equation II.10 to the expression below from Rohsenow (20).

\[
(P^*_v-P_1) = \frac{2\sigma}{r^*} - P_g
\]

The effect of the gas is to reduce the vapor pressure requirement inside the critical nucleus for a nucleus of a particular size and, therefore, reduces the superheat required for this critical nucleus. In the equations of chapters II and III the quantity \((P^*_v-P_1)\) can be thought of as the total pressure difference between the interior of the nucleus and the surrounding system. If non-condensable gas is present in the nucleus, the total pressure difference becomes \((P^*_v+P_g)-P_1\). Therefore, wherever the pressure differences occurs in these general equations it should be replaced by \((P^*_v+P_g)-P_1\).

If one can determine the mass of gas in the embryo, the pressure of that gas can be obtained from the perfect gas law.
The problem is one of determining $w_g$, the mass of gas in the embryo. Tipton (21) gives the yields of $H_2$ gas for different types of ionizing radiations in water per 100 ev of deposited energy. For fission fragments the yield of $H_2$ gas, $G(H_2)$, is approximately 1.8 molecules per 100 ev of deposited energy. The total number of molecules of $H_2$ in each embryo would be equal to the energy deposited for the embryo in ev times this yield.

$$N_g = N_{H_2} = \Delta E \cdot G(H_2) \cdot 10^6$$  \hspace{1cm} V.15

Then:

$$w_g = \frac{M_g}{g_{NaV}} \cdot \frac{M_g}{NaV} \cdot \Delta E \cdot G(H_2) \cdot 10^6$$  \hspace{1cm} V.16

where $M_g$ is the molecular weight of the gas.

Equation V.14 takes the following form when the volume of the gas, which equals the volume of the embryo, is expressed in terms of $x(i)$ and the critical radius and equation V.16 is introduced.
\[ \frac{P_g}{4\pi r^* x(1)^3} = \frac{3kT_v}{\Delta E G(H_2)} \times 10^6 \quad \text{V.17} \]

This equation is not as simple as it appears because equation V.13 make \( r^* \) a function of \( P_g \) in addition to \( T_1 = T_v \) and \( P_1 \). Also since \( E(s_2) \) depends on \( r^* \) in equation V.11, \( \Delta E \) is also a function of \( P_g \). Since \( \Delta E \) cannot be expressed explicitly in terms of \( r^* \), \( P_g \) cannot be calculated explicitly from equation V.17 and an iterative process is necessary. The procedure would go as follows for any set of variables, \([a, P_1, T_1, x(i)]\):

1. Select a value of \( P_g \)
2. Calculate \( r^* \) using equation V.13
3. Calculate \( E(s_2) \) using equation V.11
4. Calculate \( \Delta E \) using equation V.12
5. Calculate \( P_g \) using equation V.17
6. Compare with value of \( P_g \) for step 1
7. If step 5 not equal to step 1, repeat steps 1 to 6

\( P_g \)-routine-F.F.

The convergence of this procedure gives the correct values of \( P_g \), \( r^* \), and \( \Delta E \) for the particular set of variables \([a, P_1, T_1, x(i)]\). Note that for the energy balance method \( x(1) \) is unity.

The energy sink effect in equation III.14 can be handled by considering the heat of reaction of the dis-
The process takes place in the following way according to Tipton (21). The heat of reaction $Q$ defined as the difference in the enthalpies of formation of the products minus the enthalpies of formation of the reactants must be calculated to give the desired numerical results.

$$2H_2O \rightarrow 2H + 2OH$$

$$H + H \rightarrow H_2 \quad \text{or} \quad 2H_2O \rightarrow H_2 + H_2O_2$$

$$OH + OH \rightarrow H_2O_2$$

The dissociation process results from the interaction of the fission fragment with the electronic structure of the water. It may be expected, then, that the process occurs essentially while the water is in the liquid state. The hydrogen formed is, of course, in the gaseous state and the hydrogen peroxide remains in the liquid state and in the liquid region. The enthalpies of formation for $H_2O$ and $H_2O_2$ should be those for the liquid state and the reaction should be considered to take place at $T_1$. The heat of reaction in equation form for this reaction from Hatsopoulos (3) is:
\[ Q = (1) \left[ h(T_1) - h^o \right] H_2 + (1) \left[ h(T_1) - h^o \right] H_2 O_2 - (2) \left[ h(T_1) - h^o \right] H_2 O \\
+ \left[ (1) h^o_{H_2} + (1) h^o_{H_2 O_2} - (2) h^o_{H_2 O} \right] \]  

In this expression the superscript \( ^o \) signifies the standard state to which the enthalpies of formation are referred. The enthalpies at this reference state are tabulated in various references, such as Rossini (22). The enthalpy differences for each reactant and product can be found using the appropriate specific heats, such as those in Lange (23) and the temperature difference from the reference state, 25\(^\circ\)C, to the system temperature. In the temperature range (250 to 400\(^\circ\)F) of interest in this work the heat of reaction varies from 89.8 to 90.8 k cal/mole \( H_2 \). It is, then, reasonable to assume \( Q \) constant with a value of about 90.3 k cal/mole \( H_2 \) in this temperature range. The plus sign indicates that this quantity of heat must be supplied to the dissociation process for each mole of hydrogen gas formed. Then, the total energy lost for all the hydrogen formed in a track length \( ar^* \) and, therefore, contained in a single embryo is:

\[ \Delta E_x = \Delta E_{\text{radiolysis}} = \frac{QN_{H_2}}{N_A v} = \frac{Q \Delta E G(H_2)}{N_A v} 10^6 \]
V.3 Theoretical Results for Energy Balance Method

In the last section the "Pg-routine-F.F." was indicated for calculating the values of $r^*$ and $\Delta E$ which are appropriate for this case of fission fragments in water. Equation V.19 gives an explicit expression for the $\Delta E_x$ term involved. Then, the analytical criterion for radiation induced nucleation under this method, equation III.14, is rewritten below in equation V.21.

The symbol $b$ in equation III.14 must be redefined for this case due to the gas pressure involved.

\[
b = \left(\frac{P* + P_g - P_l}{P_v}\right) = \frac{2}{r^*P_\text{v}} \quad \text{V.20}
\]

Substituting equation V.20 into equation III.14 gives

\[
\frac{4}{3} \pi r^3 \left(\frac{\sigma}{r^*} + \rho_v h_{fg}\right) = \Delta E_{\text{max}} \left[1 - Q \frac{G(H_2)}{10^6 N_A}\right] \quad \text{V.21}
\]

this basic equation of the energy balance method. The left hand side is the required energy for the formation of a nucleus and the right hand side is the maximum energy available for the process. When this equation is satisfied the conditions for radiation induced nucleation have been met. Analytical expressions for all the properties in-
volved in equation V.21 and in the "P\_g-routine-F.F." as functions of temperature are given in Appendix A. Then, the combination of the "P\_g-routine-F.F." and equation V.21 has three variables involved, a, P\_1, T\_1, of which "a" and P\_1 will be considered independent.

It is obvious that the theoretical superheat threshold cannot be calculated explicitly and iterative procedures must be used. For calculational purposes another procedure called the "Sup-routine-E.B.M." will be described for calculating the superheat.

1. Select an initial value of T\_1 for a particular set (a, P\_1)
2. Feed initial value of T\_1 into "P\_g-routine-F.F." with the same set (a, P\_1)
3. Using \( \Delta E \) and \( r^* \) from the "P\_g-routine-F.F." calculate LHS and RHS of equation V.21
4. If equation V.21 is not satisfied select a new value of T\_1
5. Feed new T\_1 into "P\_g-routine-F.F." with same (a, P\_1)
6. Repeat steps 3 through 5 until equation V.21 is satisfied.

**Sup-routine-E.B.M.**

This routine gives the threshold liquid temperature or the threshold superheat for any set of variables, (a, P\_1). The complete calculational procedure is shown diagramatically in figure V.1 where the dotted lines indicate internal interactive procedures.
Figure V.1 Calculational Procedure for Obtaining the Threshold Superheat as a Function of "a" and Liquid Pressure for the Energy Balance Method.
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Table V.1 The Superheat Threshold as a Function of Pressure and the Constant "a" from the Energy Balance Method for Fission Fragments in Water.
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Table V.1 Continued.
Figure V.2  Superheat Threshold as a Function of Pressure and "a" from the Energy Balance Method for Water Exposed to Fission Fragments.
It is a simple matter to computerize this procedure and to generate a family of curves of the threshold superheat as a function of pressure with "a" as a parameter. These results are shown in table V.1 and figure V.2.

V.4 Theoretical Results for the Statistical Model

The primary equation for determining the threshold superheat in this approach is II.72. The equation is made up of two terms. The first term on the right hand side is associated with the number of nuclei formed as a result of radiation being present and the second term is associated with the number of nuclei formed completely through statistical or single molecule additions. Physically one would expect this second term to be very small compared to the first if radiation induced nucleation is observed under conditions far removed from the critical point of the substance. It is only at temperatures near this critical point that the phenomenon of purely homogeneous boiling occurs. Since this work deals with system conditions that are far from the critical point, the second term is negligible. This can also be shown through numerical comparisons.
Also involved in equation II.72 is a quantity \( X(n_R) \) which is the number of embryo os size \( n_R \) which are formed in each radiation track. Since the energy deposition rate of the fission fragment decreases along the track, each embryo which forms along the track will probably be successively smaller away from the fission site. Then it is probably reasonable to assume that only the first embryo or the embryo nearest the fission site will contribute significantly to the threshold determination. Then \( X(n_R) \) will be set equal to unity and the resulting equation for determining the threshold conditions is:

\[
\text{RRR} \cdot V_s \cdot \gamma = 1 \quad \text{V.22}
\]

In this equation there are actually seven variables, substance, radiation, radiation reaction rate, system volume, observation time, system pressure, and liquid temperature. All of these except temperature are independent variables. As far as equation V.22 is concerned the three variable, RRR, \( V_s \), and \( \gamma \) can be treated as one since they simply form a coefficient to the terms involving the dependent variable. Therefore, the following definition will be made.

\[
\text{RRR} \cdot V_s \cdot \gamma \equiv \mathcal{S} \quad \text{V.23}
\]
Now for a particular substance and type of radiation the threshold superheat depends only on the system variable and the system pressure. In this case the dimensionless effective track length "a" will also be carried as a parameter. In general, then, a set of curves of threshold superheat as a function of pressure with "a" as a parameter could be generated for each value of . Each of these sets would represent the threshold superheat for a variety of radiation reaction rates, volumes, and observation times.

A general calculational procedure will be developed for the statistical approach as was done in the preceding section. In addition to taking care of the hydrogen gas pressure and determining the threshold superheat there is the problem of determining the size of the embryos formed in the radiation tracks. The "Pg-routine-F.F." can again be used to calculate the gas pressure but this time with , the relative size of the embryo, as a necessary input. The "xR-routine" given below will handle the task of determining the embryo size generated in the radiation tracks.

1. Provide an initial value of to the "Pg-routine-F.F."

2. Receive from the"Pg-routine-F.F." and for a particular set of inputs (, P, T, )

3. Calculate from equation III.15 with equation V.19 for .
a. Calculate property values from Appendix A
b. Calculate b from equation V.20
c. Reduce equation III.15 to a function of x using steps 2, 3a, and 3b
d. Use iterative procedure to obtain x accurate to 0.0000001

4. Compare values of x from step 3d and that used in step 2

5. If x's agree to within 0.0000001, feed x = x_R to Sup-routine

6. If x's don't agree, feed x from step 3d back into "Pg-routine-F.F." at the same (a, P_1, T_1)

7. Repeat steps 2 through 6 until step 5 is satisfied

The combination of the "Pg-routine-F.F." and "x_R-routine" provide a value of x_R for any set of variables (a, P_1, T_1).

Now by holding "a" and P_1 fixed and feeding x_R in to a routine for determining the value of T_1 which fulfills the criterion for radiation induced nucleation, the procedure will be complete. The final routine is again called a "Sup-routine-S.M." and it goes as follows.

1. Select an appropriate initial value for T_1 for a particular set (a, P_1)

2. Feed this value of T_1 into the "Pg-routine-F.F."

3. Receive from the "Pg and x_R routines" the value of x_R for the set (a, P_1, T_1)

4. Calculate I_1(n_R) from equation II.61
   a. Redefine I_1(n_R) for computational purposes as
\[
I_1(n_R) = e^{-\frac{-\Delta F(n_R)}{kT_1}} \int_{x(n_R)}^{1} \frac{1}{x} \left[ 3(1-b)+2b/x(n) \right] e^{\frac{1}{kT_1} \left[ \Delta F(x(n)) - \Delta F(n_R) \right]} dx
\]

b. Calculate \( \Delta F(n_R) \) from equation II.56

c. Reduce integrand to a function of \( x \) using property value from Appendix A, \( b \) from equation V.20, and step 4b

d. Carry out integration numerically using Simpson's Rule with \( \Delta x \approx 0.00001 \)

5. Calculate \( I_2(n_R) \) from equation II.67

a. Redefine \( I_2(n_R) \) for computational purposes as:

\[
I_2(n_R) = e^{-\frac{-\Delta F(n_R)x(n_R)}{kT_1}} \int_{m}^{\infty} \left[ 3(1-b)+2b/x(n) \right] e^{\frac{1}{kT_1} \left[ \Delta F(x(n)) - \Delta F(n_R) \right]} dx/kT_1
\]

b. Use integrand from step 4c above.

c. Let \( m = x_{\min} = \text{value of } x \text{ below which the integrand is essentially zero} \).

d. Carry out integration numerically using Simpson's Rule with \( \Delta x = 0.00001 \)

6. Calculate the left hand side of equation V.22 for a particular value of \( \delta \) and \( I_1(n_R) \) and \( I_2(n_R) \) from above

7. If step 6 compares sufficiently well with unity the threshold value of \( T_1 \) for the set \( (a, P_1, \delta) \)

and, therefore, the threshold superheat is determined.
8. If not step 7, determine an appropriate new value of \( T_1 \) and feed it back into the "Pg-routine-F.F." for the same set \( (a, P_1, \mathcal{A}) \).

9. Repeat steps 3 through 8 until step 7 is satisfied.

**Sup-routine-S.M.**

The complete calculational procedure is shown schematically in figure V.3. Again the dotted line indicate interval iterative procedures.

Before applying this procedure to the specific case of fission fragments in water for the system considered in the experimental work, and interesting feature of this statistical approach will be indicated. It has to do with the dependence of the superheat threshold on the combined system variable \( \mathcal{A} \). Two sets of calculations were made to find the superheat threshold as a function of \( \mathcal{A} \) for \( a = 6 \) with \( P_1 = 14.7 \) psia in the first case and 360 psia in the second case. \( \mathcal{A} \) was allowed to vary from 10 up to \( 10^{23} \). The results are tabulated in table V.2 and plotted in figure V.4. Notice that in both cases a change in \( \mathcal{A} \) of 22 orders of magnitude resulted in changes in the superheat threshold of only a few hundredths of a degree. This indicates that for fission fragment induced nucleation of water in this pressure range the threshold superheat is essentially independent of system volume, observation time, and radiation reaction rate. This also indicates that the assumption concerning \( X(n_R) \), i.e. \( X(n_R) = 1 \), is, indeed,
acceptable since it doesn't influence the threshold conditions to any significant degree whether it is 1 or 100. This peculiarity comes about because of the nature of the integral $I_1(n_R)$ as redefined in the "Sup-routine S.M." $I_1(n_R)$ is very large in magnitude except precisely at the threshold conditions.

The particular case which is pertinent to the experimental system will now be treated using this statistical approach. Although the value of $\mathcal{S}$ is unimportant as far as the calculated superheat thresholds are concerned, a value can be assigned to it. In chapter IV it was indicated that a total reaction rate of approximately 1/minute was obtained in the system. If an observation time of about 1 minute is considered which is consistent with a temperature ramp of 1°F/min and a desired accuracy of about 1°F in the measured superheat threshold, the value of $\mathcal{S}$ is unity. Then, using the computational procedure given above, two sets of theoretical threshold superheats versus pressure were generated. The first is for $a = 6$ and the second is for $a = 10$. This will be sufficient range of theoretical values to compare with the values obtained from the energy balance method and with the experimental data. This comparison will be made in chapter VII. The theoretical results for the statistical method are tabulated in table V.3.
Figure V.3 Calculational Procedure for Calculating the Threshold Superheat by the Statistical Method for Particular Values of $\beta$, $a$, $P_1$. 

$T^*$ or $\Delta T_{\text{sup}}(a, P_1, \beta)$
<table>
<thead>
<tr>
<th>Liquid Pressure</th>
<th>T&lt;sub&gt;sat&lt;/sub&gt;(P&lt;sub&gt;1&lt;/sub&gt;)</th>
<th>Vapor Pressure</th>
<th>Gas Pressure</th>
<th>(dE/ds)&lt;sub&gt;avg&lt;/sub&gt;</th>
<th>ΔE</th>
<th>T&lt;sub&gt;sup&lt;/sub&gt;</th>
<th>ΔT&lt;sub&gt;sup&lt;/sub&gt;</th>
<th>$\mathcal{S}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lbf/ft&lt;sup&gt;2&lt;/sup&gt;</td>
<td>Deg F</td>
<td>Lbf/ft&lt;sup&gt;2&lt;/sup&gt;</td>
<td>Lbf/ft&lt;sup&gt;2&lt;/sup&gt;</td>
<td>Mev/cm</td>
<td>Mev</td>
<td>Deg F</td>
<td>Deg F</td>
<td></td>
</tr>
<tr>
<td>2116.2</td>
<td>212.000</td>
<td>5861.0</td>
<td>97.2</td>
<td>5.26(10&lt;sup&gt;4&lt;/sup&gt;)</td>
<td>17.97</td>
<td>267.999</td>
<td>55.999</td>
<td>10&lt;sup&gt;0&lt;/sup&gt;</td>
</tr>
<tr>
<td>2116.2</td>
<td>212.000</td>
<td>5857.0</td>
<td>97.0</td>
<td>5.26(10&lt;sup&gt;4&lt;/sup&gt;)</td>
<td>17.99</td>
<td>267.958</td>
<td>55.958</td>
<td>10&lt;sup&gt;6&lt;/sup&gt;</td>
</tr>
<tr>
<td>2116.2</td>
<td>212.000</td>
<td>5855.3</td>
<td>96.9</td>
<td>5.26(10&lt;sup&gt;4&lt;/sup&gt;)</td>
<td>18.00</td>
<td>267.940</td>
<td>55.940</td>
<td>10&lt;sup&gt;12&lt;/sup&gt;</td>
</tr>
<tr>
<td>2116.2</td>
<td>212.000</td>
<td>5853.8</td>
<td>96.8</td>
<td>5.26(10&lt;sup&gt;4&lt;/sup&gt;)</td>
<td>18.01</td>
<td>267.925</td>
<td>55.925</td>
<td>10&lt;sup&gt;18&lt;/sup&gt;</td>
</tr>
<tr>
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<td>212.000</td>
<td>5852.9</td>
<td>96.7</td>
<td>5.26(10&lt;sup&gt;4&lt;/sup&gt;)</td>
<td>18.01</td>
<td>267.915</td>
<td>55.915</td>
<td>10&lt;sup&gt;23&lt;/sup&gt;</td>
</tr>
<tr>
<td>51480.0</td>
<td>434.400</td>
<td>57023.7</td>
<td>874.7</td>
<td>4.91(10&lt;sup&gt;4&lt;/sup&gt;)</td>
<td>6.02</td>
<td>443.376</td>
<td>8.976</td>
<td>10&lt;sup&gt;0&lt;/sup&gt;</td>
</tr>
<tr>
<td>51480.0</td>
<td>434.400</td>
<td>57008.0</td>
<td>868.4</td>
<td>4.91(10&lt;sup&gt;4&lt;/sup&gt;)</td>
<td>6.05</td>
<td>443.349</td>
<td>8.949</td>
<td>10&lt;sup&gt;6&lt;/sup&gt;</td>
</tr>
<tr>
<td>51480.0</td>
<td>434.400</td>
<td>57001.0</td>
<td>865.6</td>
<td>4.91(10&lt;sup&gt;4&lt;/sup&gt;)</td>
<td>6.05</td>
<td>443.337</td>
<td>8.937</td>
<td>10&lt;sup&gt;12&lt;/sup&gt;</td>
</tr>
<tr>
<td>51480.0</td>
<td>434.400</td>
<td>56991.7</td>
<td>861.9</td>
<td>4.91(10&lt;sup&gt;4&lt;/sup&gt;)</td>
<td>6.07</td>
<td>443.321</td>
<td>8.921</td>
<td>10&lt;sup&gt;23&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

Table V.2. Variation of the Superheat Threshold from the Statistical Method with the System Variable, $\mathcal{S}$, for "a" = 6.0.
Figure V.4 Variation of the Superheat Threshold from the Statistical Method with the System Variable, $\mathcal{S}$, for $a = 6.0$. 

- $P_1 = 14.7$ psia
- $P_1 = 360$ psia
<table>
<thead>
<tr>
<th>Liquid Pressure Lbf/ft²</th>
<th>T_{sat}(F) Deg F</th>
<th>Vapor Pressure Lbf/ft²</th>
<th>Gas Pressure Lbf/ft²</th>
<th>(dE/ds)_{avg} Mev/cm</th>
<th>ΔE Mev</th>
<th>T_{sup} Deg F</th>
<th>ΔT_{sup} Deg F</th>
<th>&quot;a&quot;</th>
</tr>
</thead>
<tbody>
<tr>
<td>2116.2</td>
<td>212.000</td>
<td>5861.0</td>
<td>97.2</td>
<td>5.26 \times 10^{14}</td>
<td>17.97</td>
<td>267.999</td>
<td>55.999</td>
<td>6.0</td>
</tr>
<tr>
<td>11520.0</td>
<td>312.030</td>
<td>16358.7</td>
<td>260.9</td>
<td>5.19 \times 10^{14}</td>
<td>11.28</td>
<td>336.880</td>
<td>24.850</td>
<td>6.0</td>
</tr>
<tr>
<td>23040.0</td>
<td>363.530</td>
<td>28364.9</td>
<td>443.8</td>
<td>5.10 \times 10^{14}</td>
<td>8.64</td>
<td>380.251</td>
<td>16.721</td>
<td>6.0</td>
</tr>
<tr>
<td>34560.0</td>
<td>397.370</td>
<td>40074.1</td>
<td>620.4</td>
<td>5.02 \times 10^{14}</td>
<td>7.25</td>
<td>410.254</td>
<td>12.884</td>
<td>6.0</td>
</tr>
<tr>
<td>46080.0</td>
<td>423.290</td>
<td>51636.6</td>
<td>793.8</td>
<td>4.94 \times 10^{14}</td>
<td>6.35</td>
<td>433.787</td>
<td>10.497</td>
<td>6.0</td>
</tr>
<tr>
<td>2116.2</td>
<td>212.000</td>
<td>4973.2</td>
<td>82.6</td>
<td>4.85 \times 10^{14}</td>
<td>36.92</td>
<td>258.198</td>
<td>46.198</td>
<td>10.0</td>
</tr>
<tr>
<td>11520.0</td>
<td>312.030</td>
<td>15247.0</td>
<td>243.0</td>
<td>4.96 \times 10^{14}</td>
<td>23.37</td>
<td>331.689</td>
<td>19.659</td>
<td>10.0</td>
</tr>
<tr>
<td>23040.0</td>
<td>363.530</td>
<td>27123.2</td>
<td>424.1</td>
<td>4.92 \times 10^{14}</td>
<td>17.98</td>
<td>376.529</td>
<td>12.999</td>
<td>10.0</td>
</tr>
<tr>
<td>34560.0</td>
<td>397.370</td>
<td>38760.1</td>
<td>599.6</td>
<td>4.87 \times 10^{14}</td>
<td>15.14</td>
<td>407.258</td>
<td>9.888</td>
<td>10.0</td>
</tr>
<tr>
<td>46080.0</td>
<td>423.290</td>
<td>50279.8</td>
<td>772.3</td>
<td>4.81 \times 10^{14}</td>
<td>13.28</td>
<td>431.251</td>
<td>7.961</td>
<td>10.0</td>
</tr>
</tbody>
</table>

Table V.3. Theoretical Superheat Thresholds from the Statistical Method as a Function of Pressure and "a" for \( S = 1.0 \).
CHAPTER VI

Application of Statistical and Energy Balance Methods to the Case of Fast Neutrons in Water

As indicated in chapter IV the neutron cannot directly produce the highly energetic region in a substance which is necessary for radiation induced nucleation. The neutron can only interact with the nucleus of an atom and one such interaction is elastic scattering. Through this process some of the kinetic energy of a neutron can be transferred to the nucleus on which it scatters, thereby, producing a charged primary knock-on atom. This primary knock-on atom can, then, deposit its kinetic energy in much the same way as a fission fragment. Figure VI.1 indicates the two body interaction which takes place in the laboratory frame of reference.

![Figure VI.1 Scattering of a Neutron by a Nucleus.](image-url)
The laws of conservation of energy and momentum provide a relationship between the initial energy of the neutron, $E_n$, the energy of the primary knock-on atom after the event, $T$, and the scattering angle in the center of mass system, $\Theta$.

If the mass of the nucleus is taken as the mass number, $A$, and the mass of the neutron is taken as unity, the expression for $T$ becomes following Glasstone (24):

$$T = \frac{1}{2}E_n(1-\gamma)(1-\cos \Theta) \quad \text{VI.1}$$

where $\gamma$ is defined as:

$$\gamma = \left(\frac{A-1}{A+1}\right)^2 \quad \text{VI.2}$$

For neutron scattering off oxygen atoms, $A$ in equation VI.2 is 16 and $\gamma = 0.78$. Equation VI.3, then, gives the energy a primary knock-on oxygen atom would obtain as a result of scattering a neutron through an angle $\Theta$. Notice that the maximum $T$ results when a "head on" collision takes place with $\Theta = 180^\circ$. This maximum energy for oxygen is 22% of the initial neutron energy.
If the hydrogen atoms are the primary knock-ons, ∇ becomes zero and the maximum energy the hydrogen atom can obtain is 100% of the initial neutron energy. One might expect hydrogen rather than the oxygen primary knock-ons to be responsible for the radiation induced nucleation phenomenon because they can have much more energy to deposit. However, as seen in the previous chapter the nucleation phenomenon requires a high concentration of energy and, therefore, a high energy deposition rate. Appendix B shows that the oxygen primary knock-on will give the highest energy deposition rate and should be expected to control the threshold conditions for fast neutron induced nucleation.

VI.1 Energy Deposition by Primary Knock-on Oxygen Atoms

The energy deposition rate for an oxygen knock-on was treated in this work with the same equations as for fission fragments, V.1, V.2, V.3 and V.4. The subscript 1 in these equations refers to the oxygen knock-on instead of the fission fragment. Combining these equations and substituting the appropriate values gives equation VI.4. Again, the first term represents the energy transferred to the electronic system of the stopping medium and the
second term represents energy transferred by nuclear elastic scattering. Also the same assumption will be made

$$\frac{dE}{ds} = \rho_1(T_1) \left[ 1.2818 \times 10^4 \ln(0.5592E) + \frac{26.7549}{E} \ln(215.1775E) \right]$$

VI.4

regarding the energy transferred to the electronic system of the water, i.e., that this energy does result in molecular motion and is, therefore, available for the nucleation process. Some comments regarding energy transport from the track by $\delta$-electrons are made in Appendix B. For primary knock-ons with energies greater than 2 Mev this electronic energy transfer mechanism predominates and the second term may be neglected.

$$\frac{dE}{ds} = \rho_1(T_1) \left[ 1.2818 \times 10^4 \ln(0.5592E) \right]$$

VI.5

The energy deposition characteristics of the oxygen knock-on is again similar to that shown schematically in figure III.1 and the equations which apply to this type of particle may be used. Assuming the knock-on energy $J$ is known, defining $\Delta s$ again as $a r^*$, and using equation VI.5, equation III.9 can be solved for $E(s_2)$ for any prescribed liquid pressure, temperature, and parameter "a". The maximum energy deposited in the interval of track length
VI.6

\[ \frac{E(s_2)}{E(s_2) - J} \int J \rho_1(T_1) 1.2818 \times 10^4 \ln(0.5592E) \, dE = \frac{J - E(s_2)}{ar^*} \]

VI.7

\[ 0.6758a \, r^*(T_1, P_1, P_g) \rho_1(T_1) 10^4 \]

\[ \frac{[J - E(s_2)]^2}{J \ln J - E(s_2) \ln E(s_2) - 1.582 \left[ J - E(s_2) \right]} \]

ar* is then:

\[ \Delta E = J - E(s_2) \text{ Mev} \]

VI.2 Effects of the Radiolysis of Water by the Primary Knock-on Oxygen

The two effects of radiolysis can be handled in this case precisely the same as in chapter V. The gas pressure in the embryos can again be obtained using the "Pg-routine" developed in the last chapter with a few modifications. The "Pg-routine-F.N." for any set of variables \([a, P_1, T_1, \ldots]\)
and for a known value of $\mathcal{J}$ becomes:

1. Select a value of $P_g$
2. Calculate $r*$ using equation V.13
3. Calculate $E(s_2)$ using equation VI.7
4. Calculate $\Delta E$ using equation VI.8
5. Calculate $P_g$ using equation V.17
6. Compare with the value of $P_g$ for step 1
7. If step 5 not equal to step 1, repeat steps 1 through 6

This procedure gives the correct values of $P_g$, $\Delta E$, and $r*$ for the variables above and a known value of $\mathcal{J}$.

The energy sink effect is the same as for fission fragments and is given by equation V.19. The value of $G(H_2)$ may differ slightly but little error will result if the same value of 1.8 is used.

VI.3 Theoretical Calculations for Fast Neutrons in Water

The calculational procedures for both the energy balance method, E.B.M., and the statistical method, S.M.,
for this case are exactly the same as used in chapter V for the fission fragments. The energy balance method again uses the calculational procedure shown in figure V.1. The "Sup-routine-E.B.M." of section V.3 is used along with the "Pg-routine-F.N."

The statistical method uses the procedure shown in figure V.3. The "Pg-routine-F.N." above is used in combination with the "XR-routine" and "Sup-routine-S.M." of chapter V to give the theoretical results. The value of \( J \) used in this case will be somewhat different but as seen in chapter V the magnitude of \( J \) has little effect on the threshold superheat. Therefore, it is taken as unity for the following calculations.

Theoretically the threshold conditions are set by the primary knock-on oxygen atoms with the maximum \( J \), which from equation VI.3 is 22% of the available neutron energy. Then, the absolute lowest threshold superheat is set by the maximum neutron energy available. If one has a system in which there is a spectrum of neutron energies, the number of neutrons with this maximum possible energy may become negligibly small. This small number of maximum energy neutrons coupled with the small probability that a neutron will suffer a head-on collision with an oxygen nucleus tends to make the absolute superheat threshold unrealistic and unobservable. In order to measure or observe this absolute threshold one would have to
conduct an experiment with a very high overall neutron flux and/or an experiment of very long duration. Herein, lies the problem discussed briefly in chapter IV concerning the primary knock-on production rate and the desirable experimental operating conditions. In a realistic experiment designed to observe the superheat threshold, the measured threshold is not the absolute threshold. Instead it is a higher threshold corresponding to primary knock-ons which occur with sufficient frequency such that at least one will occur during the experimental time interval. This elevated threshold comes about because these participating primary knock-ons have less than the maximum energy since the production rate of knock-ons of energy $J$ is a decreasing function of $J$, i.e., higher production rate requires lower $J$. Then, if one wishes to calculate theoretical threshold superheats to compare with particular experimental values, the experiment must be analyzed to determine the actual participating primary knock-on energy. The generation and presentation of theoretical results for this case will be deferred until the experiment analysis has been performed to find the appropriate value for $J$. 


VI.4 Theoretical Development of the Participating Knock-on Energy as a Function of the Experimental Temperature Ramp

The first step in the development will be to obtain a differential production rate of primary knock-ons with energy between $I$ and $I + dI$. Two effects must be taken into account. First is the differential scattering probability or the probability that a neutron of energy $E_n$ will scatter into an angle between $\Theta$ and $\Theta + d\Theta$, thereby, imparting to the primary knock-on an energy between $I$ and $I + dI$. Second is the effect of the neutron flux spectrum, $\phi(E_n)$. The differential scattering cross-section (cross-section being the name given to reaction probabilities) per unit solid angle is given in graphical form by Goldberg (25) for neutron interaction with different elements. This cross-section may be represented as follows where $\Theta$ is again the scattering angle in the center of mass system, $\sigma'$ is the cross-section, and $\Omega'$ is the solid angle.

$$\frac{d\sigma'}{d\Omega'} = f(\cos \Theta) \text{barns/steradian} \quad \text{VI.9}$$

The term barn is a unit of cross-section and is defined as $10^{-24} \text{cm}^2$. It is desired to know the differential scatter-
ing cross-section for obtaining a knock-on of energy \( \mathcal{J} \) between \( \mathcal{J} \) and \( \mathcal{J} + d\mathcal{J} \). This can be done utilizing the "chain rule" of differential calculus.

\[
\frac{d\sigma'}{d\Omega'} = \frac{d\sigma'}{d\Omega} \frac{d\Omega}{d\sigma} \frac{d\sigma}{d\mathcal{J}}
\]

Equation VI.9 gives the expression for \( d\sigma'/d\Omega' \), \( d\Omega'/d\sigma \) is equal to \( 2\pi \sin \sigma \), and \( d\sigma/d\mathcal{J} \) can be obtained by differentiating equation VI.1. The resulting expression for the differential cross-section for producing knock-ons of energy between \( \mathcal{J} \) and \( \mathcal{J} + d\mathcal{J} \) is:

\[
d\sigma' = f(\cos \sigma) \frac{2 d\mathcal{J}}{E(1-\gamma)\sin \sigma} = \frac{4\pi f(\cos \sigma)}{E_n(1-\gamma)} d\mathcal{J}
\]

The \( \sigma \)-dependence can be replaced by solving equation VI.1 for \( \cos \sigma \) in terms of \( E_n \) and \( \mathcal{J} \). The desired differential cross-section then becomes:

\[
d\sigma'(E_n, \mathcal{J}) = \frac{4\pi g(E_n, \mathcal{J})}{E_n(1-\gamma)} d\mathcal{J}
\]

\( d\sigma'(E_n, \mathcal{J}) \) is the probability that a neutron of energy \( E_n \) will produce a primary knock-on of energy between \( \mathcal{J} \)
Figure VI.2 Relative Neutron Flux Spectrum of PuBe Neutron Sources.
heat necessitated the reduction of the water drop size from 1/2 inch in diameter to about 3/8 inch in diameter. It was found that a smaller drop could be heated to a high superheat more consistently than the larger one. This gave a system volume of 0.45 cm$^3$. The data taken for the threshold superheat as a function of ramp rate as shown in Table IV.5 indicates that the liquid temperature over the experimental range did not vary more than about 11°F. Then, it is reasonable to assume a constant liquid density of about 0.85 gm/cm$^3$ for the temperature of about 400°F. This gives a constant oxygen atom density equal to the molecular atom density (one oxygen atom per molecule of water).

\[
N_2 = N = \frac{\rho(T_1)A_Y}{M} = \frac{0.85(6.023)(10^{23})}{18} = 0.0284(10^{24})/cm^3
\]

The total neutron flux at the drop was calculated in section IV.5 as $0.53(10^4)$ neutron/cm$^2$sec. This represents the neutron flux assuming no attenuation due to the aluminum boiling chamber, insulation, and silicone oil (see figures IV.1 and IV.2). If one considers a beam of neutron from the sources to the drop, any interaction which may occur
Equation VI.14 can be integrated for each of the neutron energy ranges of equation VI.15 giving the results shown in equation VI.18. The value of $\phi$ for oxygen and the maximum neutron energy $E_{no}$ for the Pu-Be spectrum have been substituted giving the general form shown in equation VI.17.

$$dPR(J) = 2.855V_s N \phi_T \int_{4.54}^{10.5} \frac{\phi_n(E_n) \, dE_n}{E_n} \text{ number/sec}$$

Then, substituting equation VI.15 into equation VI.17 and defining $C_1$ as $V_s N \phi_T$ gives:

$$dPR(J)_1 = C_1(0.863J - 1.995lnJ - 0.3232)dJ$$

$$2.20 \leq J \leq 2.31 \text{ Mev}$$

$$dPR(J)_2 = C_1(0.0771 - 0.0950lnJ)dJ$$

$$1.98 \leq J \leq 2.20 \text{ Mev}$$

$$dPR(J)_3 = C_1(0.602J - 1.285lnJ - 0.2998)dJ$$

$$1.78 \leq J \leq 1.98 \text{ Mev}$$

To complete the evaluation of the differential production rate the coefficient $C_1$ must be evaluated. In the fast neutron experiments the requirement of higher super-
The relative neutron flux spectrum for the Pu-Be neutron sources used in the experimental program is given in figure VI.2 from Stewatt (14). The spectrum from 8 to 10.5 Mev can be represented in terms of a normalized flux $\phi_n(E_n)$ by the following function:

$$
\begin{align*}
\phi_n(E_n)_1 &= -0.0464E_n + 0.451 & 8 \leq E_n \leq 9\text{ Mev} \\
\phi_n(E_n)_2 &= 0.0333 & 9 \leq E_n \leq 10\text{ Mev} \\
\phi_n(E_n)_3 &= -0.0667E_n + 0.700 & 10 \leq E_n \leq 10.5\text{ Mev}
\end{align*}
$$

The normalized flux is obtained by dividing the relative magnitude of the flux at $E_n$ by the total flux from the sources. The flux appearing in equation VI.14 can be replaced by the normalized fluxes given by equation VI.15 multiplied by the total flux $\phi_T$.

$$
\phi_n(E_n) = \phi(E_n) \left/ \int_{\text{spectrum}} \phi(E_n) \, dE = \frac{\phi(E_n)}{\phi_T} \right.
$$

The differential scattering cross-section in angle, equation VI.9, in the range of neutron energy and scattering angle of interest here is nearly constant as shown in Goldberg (25) and has a value of approximately $50(10^{-3})b$. 
and $\mathcal{J} + d\mathcal{J}$. Then, the total production of knock-ons of this energy in a volume $V_s$ with atom density $N$ considering all capable neutrons would be the integral over all neutrons of sufficient energy to impart the energy $\mathcal{J}$. The lower limit on neutron energy would be that energy which would just impart $\mathcal{J}$ in a head-on collision. From equation VI.1 this would be:

$$\mathcal{J} = E_n (1-\gamma) \text{ or } (E_n)_{\text{min}} = \frac{\mathcal{J}}{(1-\gamma)}$$  \hspace{1cm} \text{VI.13}$$

The upper limit of the integral would be the maximum neutron energy in the spectrum, $E_{no}$. The total production rate of knock-ons of energy between $\mathcal{J}$ and $\mathcal{J} + d\mathcal{J}$ or the differential production rate, $dPR(\mathcal{J})$ is:

$$\frac{dPR(\mathcal{J})}{d\mathcal{J}} d\mathcal{J} = dPR(\mathcal{J}) = \int V_s N \sigma (E_n, \mathcal{J}) \phi(E_n) dE_n$$

$$= \frac{4 \pi}{(1-\gamma) V_s N} \int g \left( \frac{E_n}{E_n}, \mathcal{J} \right) \frac{\phi(E_n)}{E_n} dE_n d\mathcal{J}$$  \hspace{1cm} \text{VI.14}$$
whether it be scattering or absorption will either decrease the energy of the neutron and deflect it out of the beam or remove it through absorption. Also any in-scattering may be neglected since these neutrons will be degraded in energy, thereby being incapable of producing high energy knock-ons. Then, a simple transmission type calculation can be made to approximate the depletion factor for the beam or flux using the total cross-sections for the materials involved. This calculation led to a depletion factor of 58%, i.e. only 42% of the flux calculated in chapter IV is useful in forming primary knock-ons with energies of interest. Combining these values for \( N, V_s \), and \( \phi_T \) gives the value of \( C_1 \) as 28.4.

If in the experiment the minimum participating knock-on energy is \( J \), then, the total rate of production of knock-ons capable of causing nucleation is the integral of the differential production rate from \( J \) to the maximum knock-on energy produced, 2.31 Mev. All the knock-ons produced with energies equal to or greater than \( J \) are capable of causing nucleation if those of energy \( J \) are considered to be participating in the nucleation process. Substituting equation VI.18 into

\[
2.31\int_{J}^{\infty} dPR(J) \text{ number/sec} \quad \text{VI.20}
\]
equation VI.20 and carrying out the integration gives:

\[
\text{PR}(> \mathcal{J})_1 = \int_{\mathcal{J}} \text{dPR}(\mathcal{J})_1
\]

VI.21

\[
\text{PR}(> \mathcal{J})_2 = \int_{\mathcal{J}} \text{dPR}(\mathcal{J})_2 + \text{PR}(> 2.20)_1
\]

\[
\text{PR}(> \mathcal{J})_3 = \int_{\mathcal{J}} \text{dPR}(\mathcal{J})_3 + \text{PR}(> 1.98)_2
\]

These equations give the frequency at which participating knock-ons of energy >\mathcal{J} occur in the system which is at a superheat ΔT for a particular pressure. Now if knock-ons of energy \mathcal{J} are participating in the nucleation process, the required system superheat that must exist can be calculated using the methods of section VI.3. Then for a particular lower limit on participating knock-on energy \mathcal{J}, the total frequency of occurrence and the required superheat can be calculated for a particular pressure and parameter "a". Qualitatively the two functions would appear as shown in figure VI.3. If these two results are cross-plotted, the knock-on energy can be eliminated
Figure VI.3 Qualitative Representation of the Total Occurrence Frequency of Knock-ons of Energy Greater Than $\mathcal{J}$ and the Required Superheat for Knock-ons of Energy $\mathcal{J}$ to be Participating.

and the total occurrence frequency would become a function of superheat and the parameter "a" again for a particular pressure. This function is shown qualitatively in figure VI.4.

An analytical expression for this function can be obtained by fitting a polynomial to these calculated values for each value of the parameter "a". Then the form of this expression would be:

$$\text{PR}(\Delta T,a) = C_1 + C_2 \Delta T + C_3 \Delta T^2 + C_4 \Delta T^3 + \ldots \text{#/time}$$
Figure VI.4 Qualitative Representation of the Occurrence Frequency as a Function of Superheat

This expression represents the mean number of participating knock-ons which should occur in the time interval of one second. Now, if the experimental system were being heated up on a temperature ramp of $T_R^{\circ F}$/time, it can be concluded that during the system or superheat temperature change of $T_R^{\circ F}$ there should be $PR(\Delta T, a)$ knock-ons available to cause nucleation. Then, by dividing equation VI.22 by the temperature ramp, an expression which results represents the mean number of participating knock-ons which should occur during a temperature rise of $1^{\circ F}$.

Equation VI.23 can also be thought of as the relative
\[ PR'(\Delta T, a, TR) = \frac{PR(\Delta T, a)}{TR} = \]
\[
\left[ C_1 + C_2 \Delta T + C_3 \Delta T^2 + C_4 \Delta T^3 + \ldots \right] / TR \quad \text{VI.23}
\]

probability of the occurrence of a participating knock-on per degree F of temperature rise at \( \Delta T \). Likewise, \( 1 - PR'(\Delta T, a, TR) \) represents the relative probability that a participating knock-on will not occur per degree of temperature rise at \( \Delta T \). Then, if one wishes to know the actual probability that no participating knock-ons are available to cause nucleation during the heating period from the absolute superheat threshold \( \Delta T_o \) to some superheat \( \Delta T \), the probability of no knock-on occurrences during a differential temperature change must be added together from \( \Delta T_o \) to \( \Delta T \). The appropriate integral is shown in equation VI.24. Then, the probability that a participating primary knock-on occurs in a temperature interval between \( \Delta T \) and \( \Delta T + d\Delta T \) is the product of the probability of no occurrences from \( \Delta T_o \) to \( \Delta T \) and the probability

\( P_{\text{no}} \) (No nucleation up to \( \Delta T \) for "a" and TR) =

\[
\int_{\Delta T_o}^{\Delta T} \left[ 1 - PR'(\Delta T, a, TR) \right] d\Delta T = \]

\( \text{VI.24} \)
of an occurrence in $d\Delta T$ at $\Delta T$. This is in effect a differential probability that nucleation will occur in $d\Delta T$ about $\Delta T$ and will be designated as shown in equation VI.25. The mean superheat, $\overline{\Delta T}$, which one would expect if an experiment were carried out many times at the same condition and for the same TR can be defined in the usual sense as shown in equation VI.26. In this expression the integrals are carried out to an upper

$$\Delta T_m = \int_{\Delta T_0}^{\Delta T_m} \Delta T \, d\rho_{yes}(\Delta T, a, TR) \left[ \int_{\Delta T_0}^{\Delta T_m} d\rho_{yes}(\Delta T, a, TR) \right]^{-1}$$

VI.26

limit $\Delta T_m$ which is the superheat at which the differential
nucleation probability becomes zero. From equation VI.25 it can be seen that this will occur when:

\[
\frac{\Delta T_m}{\Delta T_o} \int_{PR'(\Delta T,a,TR)} \frac{d\Delta T}{\Delta T} = \Delta T_m - \Delta T_o \quad \text{VI.27}
\]

This equation can be put in integrated form using the empirical expression VI.23. \( \Delta T_m \) can then be found by a trial and error procedure on equation VI.28. Once this number is known the integrations in equation VI.26 can

\[
c_1(\Delta T_m - \Delta T_o) + \frac{c_2}{2}(\Delta T_m^2 - \Delta T_o^2) + \frac{c_3}{3}(\Delta T_m^3 - \Delta T_o^3) + \ldots = TR(\Delta T_m - \Delta T_o) \quad \text{VI.28}
\]

be performed using equation VI.25 and the empirical expression for \( PR'(\Delta T,a,TR) \) given by equation VI.23. With equation VI.26 theoretical curves of the mean expected threshold superheat for a particular value of "a" as a function of temperature ramp can be generated. A comparison of these theoretical results can be made with the experimental data given in table IV.5 to see if the calculational method is satisfactory. If so,
the results for $\overline{\Delta T}(\text{TR},a)$ can be cross plotted with the calculated results for $\Delta T(\mathcal{J},a)$ to give the final desired result which is the minimum participating primary knock-on energy as a function of the experimental procedure, $\mathcal{J}(\text{TR})$.

Two more calculational routines will now be set up to better show the necessary procedure. First, the "PR-routine" gives the production rate of primary knock-ons of energy $\mathcal{J}$ for a defined system and flux spectrum with the corresponding angular dependence of the scattering cross-section.

1. Obtain neutron flux spectrum in analytical form as equation VI.15 for PuBe sources.
2. Obtain angular dependence of scattering cross-section in analytical form as equation VI.9.
3. Calculate the atom density of primary knock-on candidates in the substance.
4. Calculate the volume of the system (it has been assumed here that the production rate is independent of position, i.e. the volume is small.).
5. Find $d\text{PR}(\mathcal{J})$ using equation VI.14.
6. Find $\text{PR}(\mathcal{J})$ using equation VI.20.
7. Calculate PR for different values of $\mathcal{J}$ using step 6.

**PR-routine**

The $\overline{\Delta T}$-routine gives the mean superheat one would expect if many experiments were performed at a temperature ramp TR for a particular "a" and $P_1$. 
1. Obtain PR'(ΔT,a,TR) from the cross-plot and curve fit of PR(ΔT) and ΔT(ΔT,a).

2. Calculate the maximum expected ΔT, Δ3T, using equations VI.27 and VI.28 with an iterative procedure.

3. Obtain ρyes from equation VI.25.

4. Calculate ΔT by numerical integration of equation VI.26 for a particular TR and "a".

**ΔT-routine**

The complete procedure for obtaining ΔT(TR) is shown in figure VI.5. Calculations of ΔT(TR,a) performed using the computer for the experimental system used in this work and at a pressure of 75 psia are shown in figure VI.6. Also shown are the experimental data of table IV.5. As can be seen fairly good agreement exists as far as the curves following the data trend. It can be assumed, then, that the computational procedure of figure VI.5 is reasonably good. Figure VI.7 shows the theoretical results for ΔT(ΔT,a) and figure VI.8 shows the calculated results for PR(ΔT). The cross plot of these two functions is shown in figure VI.9. The final cross plot of ΔT(ΔT,a) and ΔT(TR,a) shown in figure VI.9. Note that the resulting function ΔT(TR) is essentially independent of "a". With this function known the fast neutron data taken at different pressures and at known temperature ramps can be compared realistically with calculated values of threshold superheat from either of the two methods, E.B.M. or S.M. The
Figure VI.5 Procedure for Obtaining the Participating Primary Knock-on Energy as a Function of the Experimental Variable TR.
Theoretical Mean Superheat as a Function of Temperature Ramp and the Comparison with Experimental Data.

Figure VI.6 Theoretical Mean Superheat as a Function of Temperature Ramp and the Comparison with Experimental Data.
Figure VI.7 Calculated Superheat Using the Energy Balance Method as a Function of Primary Knock-on Energy.
Figure VI.8 Calculated Production Rate of Primary Knock-ons with Energy Greater Than $\mathcal{J}$ for the Experimental System.
Figure VI.9 Expected Production Rate of Primary Knock-ons at a Particular Superheat.

\[ P_1 = 75 \text{ psia} \]

Water in the Experimental System Exposed to PuBe Neutrons

\[ a = 12, 11, 10, 9, 8 \]
Figure VI.10 Theoretical Minimum Participating Primary Knock-on Energy as a Function of the Experimental Temperature Ramp.
theoretical values are now of the form $\Delta T(P_1,a,TR)$.
The actual comparison will be made in the next chapter.

VI.5 Theoretical Results for the Energy Balance and Statistical Methods

Table IV.4 indicates that the experimental temperature ramps throughout the data are in the neighborhood of $\frac{1}{2}$ degree per minute. The theoretical results will, then, be based on this TR which from figure VI.10 corresponds to a minimum participating knock-on energy of 2.12 Mev. The results for the energy balance method are shown in table VI.1 for the pressure range 14.7 psia to 360 psia and "a" values from 6 to 12. The results for the statistical method are shown in table VI.2 for the extreme values of pressure, 14.7 and 360 psia, and the extreme values of "a", 6 and 12.
<table>
<thead>
<tr>
<th>Liquid Pressure Lbf/ft²</th>
<th>T_{\text{sat}}(P_1) \text{ Deg F}</th>
<th>Vapor Pressure Lbf/ft²</th>
<th>Gas Pressure Lbf/ft²</th>
<th>(dE/\text{ds})_{\text{avg}} \text{ Mev/cm}</th>
<th>\Delta E \text{ Mev}</th>
<th>T_{\text{sup}} \text{ Deg F}</th>
<th>\Delta T_{\text{sup}} \text{ Deg F}</th>
<th>&quot;a&quot;</th>
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<td>2116.2</td>
<td>212.000</td>
<td>31765.5</td>
<td>514.3</td>
<td>2.01(10^{3})</td>
<td>0.063</td>
<td>389.830</td>
<td>177.830</td>
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<td>669.9</td>
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<td>0.057</td>
<td>414.579</td>
<td>102.549</td>
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<td>0.054</td>
<td>427.010</td>
<td>85.760</td>
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Table VI.1. The Superheat Threshold as a Function of Pressure and the Constant "a" from the Energy Balance Method for Fast Neutrons in Water.
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<th>Liquid Pressure Lbf/ft²</th>
<th>T_{sat}(P_l)</th>
<th>Vapor Pressure Lbf/ft²</th>
<th>Gas Pressure Lbf/ft²</th>
<th>(dE/ds)_avg Mev/cm</th>
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<th>T_{sup} Deg F</th>
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| 2116.2                | 212.000     | 26981.6               | 438.0               | 1.80(10³)         | 0.105 | 376.096     | 164.096        | 9.0 |
| 5760.0                | 267.250     | 31091.0               | 500.7               | 1.81(10³)         | 0.100 | 387.996     | 120.746        | 9.0 |
| 11520.0               | 312.030     | 37370.2               | 595.9               | 1.82(10³)         | 0.093 | 404.002     | 91.972         | 9.0 |
| 17280.0               | 341.250     | 43459.4               | 687.9               | 1.82(10³)         | 0.088 | 417.636     | 76.386         | 9.0 |
| 23040.0               | 363.530     | 49429.2               | 777.9               | 1.83(10³)         | 0.084 | 429.634     | 66.104         | 9.0 |
| 28800.0               | 381.790     | 55295.0               | 866.1               | 1.83(10³)         | 0.080 | 440.378     | 58.588         | 9.0 |
| 34560.0               | 397.370     | 61095.9               | 953.2               | 1.83(10³)         | 0.077 | 450.171     | 52.801         | 9.0 |
| 40320.0               | 411.050     | 66828.7               | 1039.3              | 1.83(10³)         | 0.074 | 459.170     | 48.120         | 9.0 |
| 46080.0               | 423.290     | 72519.2               | 1124.7              | 1.83(10³)         | 0.071 | 467.534     | 44.244         | 9.0 |
| 51480.0               | 434.400     | 77811.8               | 1204.4              | 1.83(10³)         | 0.069 | 474.873     | 40.473         | 9.0 |

Table VI.1. The Superheat Threshold as a Function of Pressure and the Constant "a" from the Energy Balance Method for Fast Neutrons in Water.
<table>
<thead>
<tr>
<th>Liquid Pressure Lbf/ft²</th>
<th>T&lt;sub&gt;sat&lt;/sub&gt;(F&lt;sub&gt;1&lt;/sub&gt;)</th>
<th>Vapor Pressure Lbf/ft²</th>
<th>Vapor Pressure Deg F</th>
<th>Gas Pressure Lbf/ft²</th>
<th>Gas Pressure Mev/cm</th>
<th>(dE/ds) avg</th>
<th>ΔE</th>
<th>T&lt;sub&gt;sup&lt;/sub&gt;</th>
<th>ΔT&lt;sub&gt;sup&lt;/sub&gt;</th>
<th>&quot;a&quot;</th>
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<td>212.000</td>
<td>25902.3</td>
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<td>1.74(10&lt;sup&gt;3&lt;/sup&gt;)</td>
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<td>427.569</td>
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<td>413.362</td>
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Table VI.1. The Superheat Threshold as a Function of Pressure and the Constant "a" from the Energy Balance Method for Fast Neutrons in Water.
<table>
<thead>
<tr>
<th>Liquid Pressure Lbf/ft²</th>
<th>T_{sat(P_l)} Deg F</th>
<th>Vapor Pressure Lbf/ft²</th>
<th>Gas Pressure Lbf/ft²</th>
<th>(dE/ds)_{avg} Mev/cm</th>
<th>ΔE Mev</th>
<th>T_{sup} Deg F</th>
<th>ΔT_{sup} Deg F</th>
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<tr>
<td>2116.2</td>
<td>212.000</td>
<td>24190.8</td>
<td>393.4</td>
<td>1.64(10^3)</td>
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<td>367.178</td>
<td>155.178</td>
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<td>1.70(10^3)</td>
<td>0.097</td>
<td>471.136</td>
<td>36.736</td>
<td>12.0</td>
</tr>
</tbody>
</table>

Table VI.1. The Superheat Threshold as a Function of Pressure and the Constant "a" from the Energy Balance Method for Fast Neutrons in Water.
<table>
<thead>
<tr>
<th>Liquid Pressure Lbf/ft²</th>
<th>$T_{\text{sat}}(P_1)$ Deg F</th>
<th>Vapor Pressure Lbf/ft²</th>
<th>Gas Pressure Lbf/ft²</th>
<th>$\langle dE/ds \rangle_{\text{avg}}$ Mev/cm</th>
<th>$\Delta E$ Mev</th>
<th>$T_{\text{sup}}$ Deg F</th>
<th>$\Delta T_{\text{sup}}$ Deg F</th>
<th>&quot;a&quot;</th>
</tr>
</thead>
<tbody>
<tr>
<td>2116.2</td>
<td>212.000</td>
<td>31765.5</td>
<td>514.3</td>
<td>2.01 ($10^3$)</td>
<td>0.063</td>
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<td>1.64 ($10^3$)</td>
<td>0.148</td>
<td>367.178</td>
<td>155.178</td>
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<tr>
<td>51480.0</td>
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<td>1160.9</td>
<td>1.70 ($10^3$)</td>
<td>0.097</td>
<td>471.136</td>
<td>36.736</td>
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Table VI.2. The Superheat Threshold as a Function of Pressure and the Constant "a" from the Statistical Method for Fast Neutrons in Water and for $\xi = 1.0$. 
Chapter VII

Comparison of Theoretical and Experimental Results

In this chapter some general comparisons will be made regarding the two theoretical approaches for determining the threshold conditions at which radiation induced nucleation should occur. Then, the theoretical results will be compared with the experimental data for both fission fragments and fast neutrons in water to see what can be concluded with respect to the dimensionless track length "a".

VII. 1 Comparison of the Energy Balance Method and Statistical Method

The calculated threshold superheats for fission fragments in water as a function of system pressure and "a" for the energy balance method and statistical method are shown in tables V.1 and V.3 respectively. One sees that the corresponding values in each table, i.e. for the same $P_1$ and "a", are exactly the same to three decimal places. Likewise, comparing the results of the two methods for fast neutrons in water as shown in tables
VI.1 and VI.2 shows no difference in the values obtained by the two methods.

The question that arises is, then, should these results from two entirely different methods agree precisely as indicated above? On the basis of physical reasoning the answer is yes. The experimental system which was used as a basis for calculating the theoretical results was such that the statistical growth process of the statistical approach was lost. Recall that the criterion for determining the threshold conditions with the statistical method is that one critical bubble be formed in the system of volume $V_s$ in a time interval $\tau$ when subjected to a radiation reaction rate, $HRR$, per unit volume. For the experimental system considered here the system variable $S$ which is the total number of participating embryos from which a critical sized bubble must be formed, was taken as unity. In other words there is only one embryo in the statistical population and the only way this one embryo can become critical is for it to be formed at nearly the critical size in the radiation track, i.e. $n_H = n^*$. If this one embryo is less than the critical size, the overwhelming probability is for it to condense. Then, for the case of a very small population of embryos from radiation tracks the statistical method degenerates to the energy balance method.

The effect of large populations or large $S$ was shown
in table V.2 and figure V.4. It is apparent that at the temperatures considered the statistical effect of a large population of embryos of size \( n_R \) is very small. Again the tendency for the embryos to collapse and condense overrides the tendency toward growth. The free energy barrier for growth to the critical size even for embryos very close to the critical size is much too large at these temperatures. The statistical approach could show a definite lowering of the superheat threshold from that predicted from the energy balance method at temperatures closer to the critical point. In this high temperature range the free energy barrier would become much smaller and the statistical growth process would be much more effective. Indeed if the superheat is sufficiently high for a particular pressure, the statistical growth process produces critical embryos without the aid of an external energy supply. This is the phenomenon of homogeneous nucleation which results even if \( \mathcal{J} \) is equal to zero, i.e. the radiation reaction rate is zero.

In chapter III the required energy of formation was formulated and was found to be a function of system pressure and superheat only. Nucleation occurs whenever this quantity of energy is supplied to or is in the possession of the material constituting the nucleus. The statistical theory in effect provides for a portion or perhaps all of this energy by supplying molecules to the formation process which are already in a vapor state.
Then the statistical process will provide an "effective" energy for embryo formation which will be designated $E_{\phi}$. If the state of the substance is such that $E_{\phi}$ is not equal to the energy of formation, additional energy must be supplied from a very concentrated source such as a nuclear radiation. The total energy available for formation can then be thought of as an energy contribution from radiation plus a contribution from statistical fluctuations. Note that the addition of these two quantities requires that the statistical contribution be provided in the region of and lifetime of the embryo formed from the radiation. This effect is taken into account in the rate equation treatment of chapter II. It would be expected that this coincidence requirement would severely limit the statistical contribution.

In table V.2 the superheat threshold for the same system pressure is seen to decrease slightly with increasing values of $\phi$. Since the only variable here is the population of radiation produced embryos, it seems reasonable to assume that the reduction in superheat is a statistical effect. Indeed a reduction in superheat constitutes an increase in the energy of formation as shown by the solid line in figure VII.1. This energy increase is given as a function of $\phi$ by equation VII.1 for a certain type of radiation and pressure. As

$$
\Delta E_{f}(\phi) = E_{f}\left[\Delta T_{\text{sup}}(\phi)\right] - E_{f}\left[\Delta T_{\text{sup}}(\phi =1)\right]
$$

VII.1
indicated above when \( \mathcal{J} = 1 \) the energy of formation must come almost entirely from the radiation. For

\[
E_f[\Delta T(\mathcal{J} = 1)] = E_R \quad \text{VII.2}
\]

\( \mathcal{J} \) much greater than unity the energy of formation can be provided by the sum of \( E_\mathcal{J} \) and \( E_R \). Then equation VII.1 becomes:

\[
\Delta E_f(\mathcal{J}) \approx (E_\mathcal{J} + E_R) - E_R = E_\mathcal{J} \quad \text{VII.3}
\]

\( E_\mathcal{J} \) can be computed for fission fragments and fast neutrons using the superheat drop calculated by the statistical method. These results are shown in figure VII.1 as the dashed curves for atmospheric pressure. Similar curves can be constructed for other pressures.

As the population \( \mathcal{J} \) increases the energy equivalent of the statistical growth process becomes greater as would be expected. The magnitude of the statistical contribution as calculated above appears to be much greater than might be expected. It must be kept in mind that the dashed curves are based on superheat differences of hundredths of degrees F and the accuracy of the entire calculation is therefore, questionable. There are several assumptions in the Volmer theory, which is the basis of the statistical theory used in this work, which make superheat calculations to this kind of accuracy somewhat unrealistic. Then, the qualitative nature of these curves
will be stressed and the magnitudes should be taken in the light suggested above.

It is not obvious that the statistical energy contribution should increase at lower superheat for the same value of $\mathcal{G}$. Qualitatively this may be explained on the basis of a volume effect. The spacial distribution of the statistical fluctuations in the density of "vapor state" molecules in the liquid can be taken as uniform except for the case of the maximum fluctuations which occur only a few times in the entire system. Then, assuming a uniform "density" of fluctuations throughout the liquid, a larger volume of liquid would be expected to derive a greater total benefit from the fluctuations than a smaller one. This is a manifestation of the coincidence effect referred to above which requires the fluctuations to take place within the volume of a radiation produced embryo. As the superheat is reduced the volume of the near critical embryos increases thereby increasing the coincidence probability. The magnitude and density of the fluctuations may decrease with decreased superheat but the volume or coincidence effect is overriding.

Consider now the qualitative effect of increasing $\mathcal{G}$ at constant superheat and for a particular pressure. Figure VII.2 shows the expected equilibrium number distribution of embryos when radiation produced embryos of size $n_R$ are fed into the statistical growth process. If
Figure VII.1 Energy Considerations of the Statistical Method.
Figure VII.2 Qualitative Effect of Increasing $\mathcal{S}$.

Superheat and Pressure Fixed

Embryo Size (molecules)

Number Density of Embryos

Homogeneous Embryo Formation

$(n_R)_2$

$(n_R)_1$

$\mathcal{S} \rightarrow \infty$

$n^*$
the system conditions are far away from the critical point, i.e. low pressure and superheat, the statistical growth is governed entirely by $\mathcal{G}$ since the homogeneous production of embryos of size $n_R$ is zero. Then if $\mathcal{J}$ is small, $\mathcal{G} = \mathcal{J}$, on figure VII.2, the statistical growth would be small and $n_R$ would have to be essentially equal to $n^*$. As $\mathcal{J}$ becomes much larger, $\mathcal{G} = \mathcal{J}_2$, the peak would become higher and the tail of the distribution would become longer.

Recall the analogy of the river and dam used in chapter II. Since the superheat is fixed, $n^*$ is fixed. Then $n_R$ can be reduced and still allow nucleation to take place. This is equivalent to requiring less energy from radiation as is also indicated in figure VII.1 for larger $\mathcal{G}$. In general as $\mathcal{G}$ increases still more, $n_R$ can become smaller, and $E_R$ can become smaller. In the limit of very large $\mathcal{G}$, $n_R$ and $E_R$ would approach zero and the distribution would appear as shown in figure VII.2 by the dashed line. In figure VII.1 this situation would be indicated by the energy of formation curve since $E_R \to 0$.

There appears to be an inconsistency here in that if $E_R$ goes to zero the radiation produced embryos cease to exist and $\mathcal{G} = 0$. It does not seem right that $\mathcal{G}$ can be zero and infinite at the same time. This problem can be resolved by considering the definition of $\mathcal{G}$. This quantity was defined as the product of the radiation reaction rate per unit volume, the system volume, and the observa-
tion time interval. If it is assumed that the radiation reaction rate is limited or fixed and the observation time is fixed, the variation in $\mathcal{J}$ must be brought about by a change in system volume. Then for $\mathcal{J} \to \infty$ the volume of the system is infinitely large. But for a system of infinite size the homogeneous embryo formation curve of figure VII.2 would approach the $\mathcal{J} \to \infty$ curve. Then at any particular superheat the limiting case of very large $\mathcal{J}$ is pure homogeneous nucleation.

At superheats slightly less than the maximum as set by pure homogeneous nucleation it is unclear exactly what the $E_\mathcal{J}$ curves do. It seems reasonable that they take the shape given by the dotted curves since the small size of the embryos from radiation would make the coincidence problem more acute. At the maximum superheat itself any embryos fed into the equilibrium distribution of embryos from the radiation would tend to increase the nucleation rate which already satisfies the criterion for determining the threshold superheat for nucleation without radiation. Then at this point the radiation energy contribution must go to zero thereby producing embryos of zero size so that the existing distribution is not disturbed. This is the reason for all the $E_\mathcal{J}$ curves intersecting the $E_r$ curve at the maximum superheat point in figure VII.1.

The concept of representing the degree of statistical embryo growth by an equivalent energy, $E_\mathcal{J}$, appears to be
consistent with the embryo distribution considerations as discussed above. If $E_\theta$ is calculated properly and accurately, this representation shows the complete picture in so far as when the statistical growth process is important. In general the statistical method is the correct approach for determining the threshold superheat for the occurrence of radiation induced nucleation for all system conditions. However, in the temperature range considerably below the critical point, i.e. $T_1 < 450^\circF$, the much simpler energy balance method gives very good results for all reasonable populations, $\theta$, and nearly exact results for very small $\theta$. In the following comparisons only the results from the energy balance method will be considered.

VII.2 Comparison of E.B.M. Results and Experimental Data for Fission Fragments in Water

The experimental data obtained in this work from table IV.2 and Dietrich's data from table IV.3 have been plotted in figure VII.3 along with the theoretical results of table V.1. The figure is split into a low and high pressure range to allow the temperature scale to be expanded. The data obtained in this work appears to follow the $a = 6$ line very closely up to about 50 psia and, then, drops gradually to a higher value of "a" as the pressure increases. Dietrich's data in the higher pressure range
Figure VII.3 Comparison of Theoretical and Experimental Superheat Thresholds for Fission Fragments in Water.
Figure VII.3 Continued at Higher Pressure.

- Data from this work with indicated precision
- Deitrich's data with indicated uncertainties (11)
Figure VII.4 Comparison of Theoretical and Experimental Threshold Superheats for Fast Neutrons in Water.
also indicates a continuation of this trend of increasing "a" with pressure but, then, reverses at about 200 psia. It is difficult to know if these trends are real or due to inaccuracies in the data. It should be noted that the deviation from a constant value of "a" for the entire range of pressures is only about 3 degrees.

VII.3 Comparison of E.B.M. Results and Experimental Data for Fast Neutrons in Water

The theoretical results of table VI.1 and the experimental data of table IV.4 are plotted in figure VII.4 for the fast neutron case. It appears that the "a" value for fast neutrons is in the neighborhood of 10. The deviation of this data from the $a = 6$ curve is on the order of 12 degrees which may or may not be greater than experimental error.

An attempt will be made in the next section to indicate the possible experimental and theoretical errors or uncertainties involved.

VII.4 Discussion of Uncertainties

There are various uncertainties both in the theoretical development and in the data which must be recognized be-
fore any definitive statements or conclusions can be made regarding the correct trend in "a". Each of these uncertainties will be explained and an estimate of their magnitudes will be given.

Consider first the problem of the correct determination of the energy deposition rate of a heavy charged particle passing through a liquid. The main uncertainty in this regard is the mean charge which the particle possesses as a function of its velocity. The theoretical superheat results are very sensitive to this factor because the mean energy deposition rates used depend on the square of the mean effective charge of the particle. Considerable work has been done on determining the charge-velocity dependence of heavy particles in gases. Nikolaev (26) considers this dependence for heavy ions such as oxygen, nitrogen, carbon, etc. in different gases and in a celluloid film. He indicates that in general the Bohr theory (equation V.2) over estimates the charge for gases. However, the experiments showed a marked increase in the mean charge of particular ion when it passed through the celluloid film instead of gas. Bell (27) indicates that the same general behavior exists for fission fragments in condensed materials as opposed to gases. The Bohr theory used in this work may be fairly realistic for predicting the mean charge of heavy particles as a function of velocity but it is not difficult to justify an uncertainty
of at least 20% in the magnitude of the mean charge. The
final curves presented in figure VII.5 and VII.6 show
this uncertainty as dashed lines.

The uncertainties discussed now will concern the
experimental data. Consider first the problem already
mentioned in chapter IV concerning the nonisothermal
condition of the experimental apparatus in the vicinity
of the water drop. A correction was developed in chapter
IV and applied uniformly to all the data. It is unrealis-
tic to claim that this correction is accurate to less
than $\pm 2^\circ F$. This number will be used as an estimate in
an attempt to determine a overall uncertainty for the
superheat data.

The thermocouple and temperature measuring system is
another source of uncertainty. The calibration at low
temperature was very good but at high temperatures, 300-
400$^\circ F$, the inaccuracy could easily be around $2^\circ F$ and may
or may not be systematic in nature. Therefore, and un-
certainty of $\pm 2^\circ F$ in superheat will be assigned to the
thermocouple and temperature measuring system.

The pressure determination is another source of un-
certainty. Although the gage was calibrated it is im-
possible to read the gage to anything less than 1/2 psi.
This would correspond to an uncertainty in the saturation
conditions and, therefore, the measured superheat. In
the pressure range considered in this work, this uncertain-
ty would not amount to more than $\pm 1^\circ F$ in superheat.

Tso (28) has pointed out that a systematic uncertainty of perhaps considerably magnitude exists in the experimental apparatus for which no correction was made. This has to due with the aluminum thermocouple probe used in making the temperature measurements. This probe extends up from the bottom of the boiling chamber through the convection generator to the bottom of the drop (see figure IV,2). Since the oil is cooled as it flows downward through the convection generator, a temperature gradient is set up along the axis of the probe. Due to the relatively thick wall design of the probe and the high thermal conductivity of the aluminum, sufficient heat was being conducted down the probe from the tip where the thermocouple bead was located to set up appreciable temperature differences from the bead to the oil outside the probe. He measured this temperature difference at low temperature, approximately $200^\circ F$, and at a convection generator flow rate about two or three times greater than that used in taking this data. The difference was about $4^\circ F$ and was found to be quite sensitive to the convection generator flow rate. It is difficult to assign an uncertainty to this effect because it will certainly not be constant with temperature. Since the convection generator operates at near atmospheric pressure, as the system operating temperature increases and more and more boiling takes
place in the convection generator a greater amount of heat is removed from the oil for a constant flow rate. Then, it would be expected that this temperature difference through the wall of the probe would become greater as the system temperature increased. Rough estimates of this uncertainty would be approximately 2°F for the low temperature fission fragment runs where little or no boiling takes place in the convection generator and perhaps 6°F for the highest temperature fast neutron runs where nearly all the convection generator flow was known to be vaporized. For these two extreme cases the heat removed by the generator increased by about a factor of 6 due to the vaporization for a constant flow in all cases.

One other uncertainty exists for the fission fragment data due to the presence of a salt in the water. The main effect of the solute would be on the surface tension. Weast (29) lists the surface tension of many inorganic aqueous solutions as a function of concentration at normal room temperature. In general all the salts tend to raise the surface tension by less than 1% for concentrations in the range used in this work. Some theoretical calculations were made to indicate the magnitude of the effect of altering the surface tension. It was found that a 1% change in surface tension would change the superheat by at most 0.5°F. In light of the other uncertainties this effect is small and can be neglected.
The uncertainties indicated above will now be combined to give an estimate of the total uncertainty in the data. The random uncertainties due to the nonisothermal condition, temperature measurement, and pressure measurement can be combined in the usual way, i.e. the square root of the sum of the squares, to give a total random uncertainty of \( \pm 3^\circ F \). To this must be added the systematic uncertainty due to the thermocouple probe. Using the rough estimates above and linearly interpolating between the extremes of approximately \( 270^\circ F \) and \( 420^\circ F \) in temperature gives a total uncertainty as a function of temperature as \([-3, + 3 + \left(\frac{1}{38} T-5\right)]\) or \([-3, + \left(\frac{1}{38} T-2\right)]\).

The data as plotted in figures VII.5 and VII.6 show the total estimated uncertainty indicated above. Tso (28) should be able to remove most of this uncertainty in his work with modification to the same apparatus and obtain data to within \( \pm 2^\circ F \).

VII.5 Theoretical Determination of "a"

There are two possible approaches to determining the appropriate value of the effective dimensionless track length "a". First it is possible to correlate the data with an empirical expression. The resulting predic-
tion of "a" could not be expected to be any better than the data on which it is based and would certainly be suspect when applied to a different substance or to considerably different conditions of the same substance. In light of the uncertainties associated with the data and the theory as discussed in the previous section little confidence should be placed in such a correlation.

A second and more satisfying approach is to base the prediction of "a" on a reasonable and fundamental physical model. If the model is sufficiently sound, it can be more confidently extended to other conditions and substances. Such a model should of course be verified by experimental data if possible. Both approaches will be developed in this section with emphasis on the model approach.

The physical basis of the proposed model is that the energy deposited by the energetic particle passing through the material, which manifests itself as molecular motion or heat in the usual sense, leads to the formation of a cylinder of vapor along the particle's track. This cylinder will grow intact to a maximum size which will then break-up into spherical bubbles. It is assumed that the growth rate of the cylinder is sufficiently rapid that all possible break-up phenomenons are inoperative during the growth phase. At the point of maximum size a relatively static state exists. It will also be assumed that the
surface of this cylinder contains a wide variety of disturbances, i.e., disturbances of all wave lengths, as a result of the rapid expansion, nonuniform energy deposition along the track, etc. Rayleigh (30) has developed a theory of "jet instabilities" which may be applied to this situation. In general a column or jet of vapor in a liquid is unstable to all surface disturbance with wave lengths greater than the circumference of the vapor column. Rayleigh (31) shows, however, that there is a certain wave length which leads to the maximum rate of growth of the disturbance amplitude. This wave length is given by equation VII.4 where \( d_c \) is the diameter of the cylinder. Since it has been assumed that many different wavelengths exist, the disturbance

\[
\lambda = 6.48 d_c \quad \text{VII.4}
\]

with this particular wave length will literally out-grow all others and be responsible for the break-up of the cylinder. This wave length is then the effective track length desired. With the definition of "a" equation VII.4 becomes:

\[
\lambda = L = ar^* = 6.48 d_c = 12.96 r_c
\]

\[ a = 12.96 \frac{r_c}{r^*} \quad \text{VII.5}
\]

If the maximum radius of the cylinder can be obtained as a function of liquid state, "a" would be given
for any pressure and superheat. This radius can be found very simply from the expression for the energy of formation of a critical cylinder. A critical cylinder is defined in the same way as a critical sphere neglecting end effects. The energy of formation can also be derived in the same manner as for a sphere and is given by equation VII.6. For a cylinder the dynamic

\[(E_f)_c = \pi r_c^2 L \rho_v h_{fg} + \pi r_c^2 L (P_v - P_1)\] VII.6

equilibrium expression gives the pressure difference as:

\[P_v - P_1 = \sigma/r_c\] VII.7

The energy of formation then becomes:

\[(E_f)_c = \pi r_c^2 L (\rho_v h_{fg} + \sigma/r_c)\] VII.8

The energy available for the formation of this cylinder is the same as for the spherical case of chapter III where \[L = ar^* = 12.96 r_c\]. Equation III.9

\[E_A = \Delta E_{\text{max}} - \sum_x \Delta E_x\] VII.9

reduces to the following for water

\[E_A = \text{Const.} \cdot (L)(dE/dS)_{\text{avg. over } L}\] VII.10

Combining equations VII.8 and VII.10 gives the expression for \(r_c\) in terms of properties and the average energy deposition rate. Using equation


\[
\text{Const. } \frac{(dE/dS)}{\text{avg.}} = \pi r_c^2 (\rho_v h_{fg} + \sigma/r_c) \text{ VII.11}
\]

VII.5 in this expression results in an expression for "a" in terms of pressure, superheat, and average energy deposition rate. The energy deposition

\[
\text{Const. } \frac{(dE/dS)}{\text{avg.}} = \frac{\pi a^2 r^*}{(12.96)^2} \left( \rho_v h_{fg} + \frac{12.96 \sigma}{a r^*} \right) \text{ VII.12}
\]

rate can be eliminated with equation III.14 which gives the criterion for spherical nucleation.

\[
\frac{4}{3} \pi r^* \left( \rho_v h_{fg} + \frac{\sigma}{r^*} \right) = \text{Const. } \frac{(dE/dS)}{\text{avg.}} a r^* \text{ VII.14}
\]

Then:

\[
\frac{4\pi r^*}{3a} \left( \rho_v h_{fg} + \frac{\sigma}{r^*} \right)_{\text{sphere}} = \frac{a^2 r^*}{(12.96)^2} \left( \rho_v h_{fg} + \frac{12.96 \sigma}{a r^*} \right)_{\text{cylinder}}
\]

\[
a = \left\{ \frac{224 (\rho_v h_{fg} + \frac{\sigma}{r^*})_{\text{sphere}}}{(\rho_v h_{fg} + \frac{12.96 \sigma}{a r^*})_{\text{cylinder}}} \right\}^{1/3} \text{ VII.15}
\]

The terms in parenthesis are subscripted because they would be evaluated at slightly different conditions. If the nucleation is to be spherical nucleation instead of cylindrical, then the cylinder from which the sphere evolves must be somewhat subcritical. Being subcritical at its maximum size means that its radius is slightly less and therefore its internal pressure slightly greater than the critical cylinder. The internal temperature in the subcritical cylinder would therefore be somewhat higher. The overall effect of the slightly subcritical cylinder
is to increase the term in the denominator of equation VII.15 above that of a critical cylinder. Calculations show this effect to be about 2% in the value of "a" for the worst case and will therefore be neglected. Calculation also show that the \( \sigma/r^* \) terms in the parenthesis are at most about 3% of \( \rho v h_{fg} J \). Neglecting these terms entirely would affect "a" by about 1%. Then even in the worst case equation VII.15 reduces to a constant value of "a" to within 3%.

This is indeed and interesting result in that "a" is equal to \( 6.07 \pm 3\%_{\text{max.}} \), independent of pressure, superheat, type of radiation, and substance. If this model of cylindrical growth and instability break-up is valid, "a" appears to be nearly a universal constant.

The data available at present suggests that the effective track length for a particular type of radiation is constant with pressure. The data of the two different types of radiation indicate a correlation with the square root of the energy deposition rate. These considerations lead to the following correlation and empirical constant.

\[
a = 4.6 \sqrt{(dE/dS)_{\text{avg.}}/r^*} \quad \text{VII.16}
\]

This dependence would indicate that the determination of the effective track length is brought about very early in the lifetime of the track. Only at the beginning of the expansion process is the external pressure always negli-
gible. Or it may suggest a phenomenon which is in some way related to the energy deposition process itself.

VII.6 Time Considerations for the Instability Model
Determination of "a"

In the previous section it is assumed that the instability induced reconfiguration of the vapor cylinder into spherical bubbles takes place while the cylinder is in a nearly static state. This static state exists only from the completion of the cylindrical growth process to the beginning of the collapse process since the cylinder is slightly subcritical. In order for this assumption to hold, thereby giving the maximum possible $L = 12.96\ r_c$, the instability growth and subsequent pinch-off must be sufficiently rapid that appreciable cylinder collapse cannot occur.

A time constant for the instability growth can be obtained following Rayleigh (31). The amplitude of the wave instability has a time dependent term as shown in equation VII.17. The time

$$\text{Amplitude} \propto e^{gt} \quad \text{VII.17}$$

constant can be defined as $t = q^{-1}$ which is the time required for the amplitude to increase by a factor of $e$. For the wave length of maximum instability this time
constant becomes:

\[ q^{-1} = 1.22 \left[ \frac{\rho r_0^3}{g_0 \sigma} \right]^{1/2} \text{ sec.} \quad \text{VII.18} \]

The dynamics of bubble collapse has been treated by Florschuetz (37) for both the inertia controlled case and the heat transfer controlled case. Then a time constant for the process can be defined as the time required for the bubble radius to decrease by a factor of e. From the graphical results given by Florschuetz the dimensionless relaxation times given in equations VII.19 and VII.20 are obtained.

\[ \gamma_{\text{Inertia controlled}} = \frac{t_1}{r} \sqrt{\frac{2 \Delta P}{3 A}} = 0.7 \quad \text{VII.19} \]

\[ \gamma_{\text{Heat transfer controlled}} = \frac{4}{\pi} Ja \frac{D_I t_H}{r^2} = 1.0 \quad \text{VII.20} \]

Ja is the Jakob number and is defined as:

\[ Ja = \frac{\rho C_\ell \Delta T}{\rho_v h_{fg}} \quad \text{VII.21} \]

The other symbols have their usual meaning with \( D_I \) and \( C_\ell \) being the thermal diffusivity and specific heat respectively. \( \Delta P \) in equation VII.19 is the increase in system pressure which brings about the collapse in the bubble. \( \Delta T \) in equation VII.20 is the change in saturation temperature due to \( \Delta P \). The Clapeyron relation can be used to relate these two quantities.

\[ \Delta P = \Delta T \frac{J_{h_{fg}} \rho_v}{T} \quad \text{VII.22} \]
Solving equations VII.19 and VII.20 for the real time constants, $t_I$ and $t_H$, and substituting equation VII.21 and VII.22 gives:

$$t_I = 0.7 \frac{r^{3/2} \Delta P g \rho_{ch}}{2} \text{ sec.} \quad \text{VII.23}$$

$$t_H = \frac{1.0 \pi r^2 h_f g \rho_v J^2}{\rho_t^2 C_v^2 T^2 \Delta P D} \quad \text{VII.24}$$

Substituting a few typical numbers into these expressions indicates that the time constant for heat transfer controlled collapse is very much larger than that for inertial collapse. This means that the ability to transfer the heat of vaporization away from the bubble controls the process. Even in the case of sodium where the heat transfer is much superior to water the $t_H$ is still greater than $t_I$ up to $\Delta P$'s of about $10^6$ lbf/ft$^2$ or about 700 psia. In the relatively low pressure range of substances this sort of pressure difference can not exist.

The important comparison to make at this point is the relative magnitudes of the time constant for vapor bubble collapse and the time constant for the growth of a surface instability on a vapor cylinder. The time constant for bubble collapse as given by equation VII.24 is for a spherical bubble and not a cylinder. However, in a heat transfer controlled situation the ratio of the heat transfer area or interfacial area to the change in vapor mass with respect to radius is the important parameter affecting the rate of collapse. For a sphere and a cylinder this
ratio is the same and estimating the heat transfer controlled relaxation time for a cylinder using equation VII.24 may not be too bad.

Typical numbers of interest in this work for sodium give \( q^{-1} \) from equation VII.18 as \( 1.5 \times 10^{-8} \) sec. For a \( \Delta P \) on the order of 100 psi the \( t_H \) from equation VII.24 for sodium under the same typical conditions is about \( 2.5 \times 10^{-6} \) sec. It seems reasonable to assume that the lifetime of the relatively static vapor cylinder is sufficiently long to allow the instability break-up phenomenon to operate before appreciable collapse occurs. For typical water conditions, \( q^{-1} = 1.5(10^{-8}) \) sec. and \( t_H \approx 4(10^{-5}) \) sec.

VII.7 Comparison of Final Results

The final calculated superheats for fission fragments in water are shown in table VII.1 for the empirical representation of "a" and tables VII.2, VII.3, and VII.4 for the "constant a" theory. Table VII.2 gives superheats based on the energy deposition rate for the mean charge predicted by the Bohr equation, equation V.2, and tables VII.3 and VII.4 give superheats for a ± 20% uncertainty in this mean charge. Figure VII.5 shows all of these calculated results along with the data of this work and Deitrich's data. The "constant a" theory certainly appears to represent the situation fairly well for fission frag-
ments in water. Deitrich's (11) data around 200 psia tends to fall about 2 degrees F below the predicted "constant a" line but then comes into agreement at higher pressure.

The calculated results for fast neutrons are tabulated in tables VII.5, VII.6, VII.7, and VII.8 for the same representations for "a" and the same mean charge variations as for the fission fragments. These results are plotted in figure VII.6 along with the fast neutron data. The agreement here is not as good as for the fission fragments but considerable overlap does exist in the uncertainties. This limited agreement, in light of the known but estimated uncertainties in the data, certainly does not disprove the "constant a" theory.
<table>
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<th>Liquid Pressure Lbf/ft²</th>
<th>T_{\text{sat}}(P_l) Deg F</th>
<th>Vapor Pressure Lbf/ft²</th>
<th>Gas Pressure Lbf/ft²</th>
<th>(dE/ds)_{avg} Mev/cm</th>
<th>ΔE Mev</th>
<th>T_{sup} Deg F</th>
<th>ΔT_{sup} Deg F</th>
<th>&quot;a&quot;</th>
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<td>111.8</td>
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<td>16.90</td>
<td>276.920</td>
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<td>16.16</td>
<td>334.190</td>
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Table VII.1 Calculated Results for Fission Fragments in Water Using the Empirical Relation, 

\[ a = 4.6 \sqrt{\text{dE/ds}} /r^* \]

for "a" and the Predicted Values of dE/ds.
<table>
<thead>
<tr>
<th>Liquid Pressure (Lbf/ft²)</th>
<th>T_{sat}(P_1) (Deg F)</th>
<th>Vapor Pressure (Lbf/ft²)</th>
<th>Gas Pressure (Lbf/ft²)</th>
<th>(dE/ds)_{avg} (Mev/cm)</th>
<th>ΔE</th>
<th>T_{sup} (Deg F)</th>
<th>ΔT_{sup} (Deg F)</th>
<th>&quot;a&quot;</th>
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Table VII.2 Calculated Results for Fission Fragments in Water Using the "Constant a" Theory and the Predicted Values of dE/ds.
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<th>Liquid Pressure ( \text{Lbf/ft}^2 )</th>
<th>( T_{\text{sat}}(P_1) ) Deg F</th>
<th>Vapor Pressure ( \text{Lbf/ft}^2 )</th>
<th>Gas Pressure ( \text{Lbf/ft}^2 )</th>
<th>( (dE/\text{ds})_{\text{avg}} ) Mev/cm</th>
<th>( \Delta E ) Mev</th>
<th>( T_{\text{sup}} ) Deg F</th>
<th>( \Delta T_{\text{sup}} ) Deg F</th>
<th>( &quot;a&quot; )</th>
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Table VII.3 Calculated Results for Fission Fragments in Water Using the "Constant a" Theory and Values of \( dE/\text{ds} \) Representing a Positive Uncertainty in the Mean Charge of 20%.
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<th>Vapor Pressure Lbf/ft²</th>
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<td>451.211</td>
<td>11.611</td>
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</tr>
<tr>
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<td>444.590</td>
<td>64602.5</td>
<td>989.4</td>
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<td>3.01</td>
<td>455.749</td>
<td>11.159</td>
<td>6.07</td>
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</table>

Table VII.4 Calculated Results for Fission Fragments in Water Using the "Constant a" Theory and Values of dE/ds Representing a Negative Uncertainty in the Mean Charge of 20%.
Figure VII.5 Comparison of Final Theories for the Superheat Threshold for Fission Fragments in Water with the Data.
<table>
<thead>
<tr>
<th>Liquid Pressure Lbf/ft²</th>
<th>T\text{sat}(P_l)\ Deg F</th>
<th>Vapor Pressure Lbf/ft²</th>
<th>Gas Pressure Lbf/ft²</th>
<th>(dE/ds)_{avg} Mev/cm</th>
<th>T\Delta E Mev</th>
<th>T\sup\ Deg F</th>
<th>ΔT\sup \ Deg F</th>
<th>&quot;a&quot;</th>
</tr>
</thead>
<tbody>
<tr>
<td>2116.2</td>
<td>212.000</td>
<td>26953.1</td>
<td>440.3</td>
<td>1.79(10³)</td>
<td>0.106</td>
<td>376.000</td>
<td>164.000</td>
<td>9.</td>
</tr>
<tr>
<td>2880.0</td>
<td>227.960</td>
<td>27698.2</td>
<td>449.4</td>
<td>1.79(10³)</td>
<td>0.106</td>
<td>378.260</td>
<td>150.300</td>
<td>9.</td>
</tr>
<tr>
<td>5760.0</td>
<td>267.250</td>
<td>30642.3</td>
<td>494.2</td>
<td>1.78(10³)</td>
<td>0.106</td>
<td>386.750</td>
<td>119.500</td>
<td>9.</td>
</tr>
<tr>
<td>8640.0</td>
<td>292.710</td>
<td>33619.2</td>
<td>544.3</td>
<td>1.78(10³)</td>
<td>0.105</td>
<td>394.710</td>
<td>102.000</td>
<td>10.</td>
</tr>
<tr>
<td>11520.0</td>
<td>312.030</td>
<td>36427.1</td>
<td>581.7</td>
<td>1.77(10³)</td>
<td>0.104</td>
<td>401.730</td>
<td>89.700</td>
<td>10.</td>
</tr>
<tr>
<td>14400.0</td>
<td>327.810</td>
<td>39273.9</td>
<td>623.9</td>
<td>1.76(10³)</td>
<td>0.104</td>
<td>408.430</td>
<td>80.620</td>
<td>10.</td>
</tr>
<tr>
<td>17280.0</td>
<td>341.250</td>
<td>42114.5</td>
<td>667.0</td>
<td>1.76(10³)</td>
<td>0.103</td>
<td>414.750</td>
<td>73.500</td>
<td>10.</td>
</tr>
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<td>353.020</td>
<td>44941.7</td>
<td>710.2</td>
<td>1.75(10³)</td>
<td>0.103</td>
<td>420.720</td>
<td>67.700</td>
<td>10.</td>
</tr>
<tr>
<td>23040.0</td>
<td>363.530</td>
<td>47745.3</td>
<td>752.3</td>
<td>1.74(10³)</td>
<td>0.102</td>
<td>426.360</td>
<td>62.830</td>
<td>11.</td>
</tr>
<tr>
<td>25920.0</td>
<td>373.060</td>
<td>50554.4</td>
<td>795.4</td>
<td>1.74(10³)</td>
<td>0.102</td>
<td>431.760</td>
<td>58.700</td>
<td>11.</td>
</tr>
<tr>
<td>28800.0</td>
<td>381.790</td>
<td>53328.6</td>
<td>835.9</td>
<td>1.73(10³)</td>
<td>0.101</td>
<td>436.870</td>
<td>55.080</td>
<td>11.</td>
</tr>
<tr>
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<td>389.860</td>
<td>56149.4</td>
<td>883.7</td>
<td>1.73(10³)</td>
<td>0.101</td>
<td>441.860</td>
<td>52.000</td>
<td>11.</td>
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<tr>
<td>34560.0</td>
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<td>918.1</td>
<td>1.72(10³)</td>
<td>0.100</td>
<td>446.490</td>
<td>49.120</td>
<td>11.</td>
</tr>
<tr>
<td>37440.0</td>
<td>404.420</td>
<td>61625.3</td>
<td>959.3</td>
<td>1.72(10³)</td>
<td>0.100</td>
<td>451.020</td>
<td>46.600</td>
<td>11.</td>
</tr>
<tr>
<td>40320.0</td>
<td>411.050</td>
<td>64378.0</td>
<td>1000.5</td>
<td>1.71(10³)</td>
<td>0.099</td>
<td>455.390</td>
<td>44.340</td>
<td>12.</td>
</tr>
<tr>
<td>43200.0</td>
<td>417.330</td>
<td>67138.2</td>
<td>1043.8</td>
<td>1.71(10³)</td>
<td>0.099</td>
<td>459.630</td>
<td>42.300</td>
<td>12.</td>
</tr>
<tr>
<td>46080.0</td>
<td>423.290</td>
<td>69865.5</td>
<td>1082.6</td>
<td>1.70(10³)</td>
<td>0.098</td>
<td>463.690</td>
<td>40.400</td>
<td>12.</td>
</tr>
<tr>
<td>48960.0</td>
<td>428.970</td>
<td>72620.7</td>
<td>1126.6</td>
<td>1.70(10³)</td>
<td>0.098</td>
<td>467.670</td>
<td>38.700</td>
<td>12.</td>
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<tr>
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<td>1165.3</td>
<td>1.69(10³)</td>
<td>0.098</td>
<td>471.490</td>
<td>37.090</td>
<td>12.</td>
</tr>
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<td>1205.8</td>
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<td>0.097</td>
<td>475.210</td>
<td>35.610</td>
<td>12.</td>
</tr>
<tr>
<td>57600.0</td>
<td>444.590</td>
<td>80799.7</td>
<td>1247.6</td>
<td>1.68(10³)</td>
<td>0.097</td>
<td>478.840</td>
<td>34.250</td>
<td>12.</td>
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</table>

Table VII.5 Calculated Results for Fast Neutrons in Water Using the Empirical Relation, $a = 4.6\sqrt{dE/ds}/r^*$, for "a" and the Predicted Values of dE/ds.
<table>
<thead>
<tr>
<th>Liquid Pressure T_{sat}(P_1)</th>
<th>Vapor Pressure</th>
<th>Gas Pressure</th>
<th>(dE/ds)_{avg}</th>
<th>ΔE</th>
<th>T_{sup}</th>
<th>ΔT_{sup}</th>
<th>&quot;a&quot;</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lbf/ft^2</td>
<td>Deg F</td>
<td>Lbf/ft^2</td>
<td>Deg F</td>
<td>Lbf/ft^2</td>
<td>Mev/cm</td>
<td>Mev</td>
<td>Deg F</td>
</tr>
<tr>
<td>2116.2</td>
<td>212.000</td>
<td>31616.9</td>
<td>512.0</td>
<td>2.01(10^3)</td>
<td>0.064</td>
<td>389.420</td>
<td>177.420</td>
</tr>
<tr>
<td>2880.0</td>
<td>227.960</td>
<td>32475.7</td>
<td>525.0</td>
<td>2.01(10^3)</td>
<td>0.064</td>
<td>391.720</td>
<td>163.760</td>
</tr>
<tr>
<td>5760.0</td>
<td>267.250</td>
<td>35677.2</td>
<td>573.7</td>
<td>2.01(10^3)</td>
<td>0.061</td>
<td>399.900</td>
<td>132.650</td>
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<tr>
<td>8640.0</td>
<td>292.710</td>
<td>38812.4</td>
<td>621.3</td>
<td>2.02(10^3)</td>
<td>0.059</td>
<td>407.370</td>
<td>114.660</td>
</tr>
<tr>
<td>11520.0</td>
<td>312.030</td>
<td>41893.4</td>
<td>667.9</td>
<td>2.02(10^3)</td>
<td>0.058</td>
<td>414.270</td>
<td>102.240</td>
</tr>
<tr>
<td>14400.0</td>
<td>327.810</td>
<td>44941.7</td>
<td>714.0</td>
<td>2.02(10^3)</td>
<td>0.056</td>
<td>420.720</td>
<td>92.910</td>
</tr>
<tr>
<td>17280.0</td>
<td>341.250</td>
<td>47944.0</td>
<td>759.2</td>
<td>2.02(10^3)</td>
<td>0.055</td>
<td>426.750</td>
<td>85.500</td>
</tr>
<tr>
<td>20160.0</td>
<td>353.020</td>
<td>50917.0</td>
<td>803.9</td>
<td>2.03(10^3)</td>
<td>0.053</td>
<td>432.440</td>
<td>79.420</td>
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<tr>
<td>23040.0</td>
<td>363.530</td>
<td>53862.6</td>
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<td>0.052</td>
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<td>373.060</td>
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<td>892.0</td>
<td>2.03(10^3)</td>
<td>0.051</td>
<td>442.950</td>
<td>69.890</td>
</tr>
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<td>447.850</td>
<td>66.060</td>
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<td>0.049</td>
<td>452.540</td>
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</tr>
<tr>
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<td>397.370</td>
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<td>457.050</td>
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<td>404.420</td>
<td>68303.5</td>
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<td>56.960</td>
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<td>411.050</td>
<td>71136.0</td>
<td>1107.7</td>
<td>2.03(10^3)</td>
<td>0.046</td>
<td>465.540</td>
<td>54.490</td>
</tr>
<tr>
<td>43200.0</td>
<td>417.330</td>
<td>73965.0</td>
<td>1150.5</td>
<td>2.03(10^3)</td>
<td>0.046</td>
<td>469.570</td>
<td>52.240</td>
</tr>
<tr>
<td>46080.0</td>
<td>423.290</td>
<td>76776.8</td>
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<td>2.03(10^3)</td>
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</tr>
<tr>
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<td>428.970</td>
<td>79579.3</td>
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<td>2.03(10^3)</td>
<td>0.044</td>
<td>477.230</td>
<td>48.260</td>
</tr>
</tbody>
</table>

Table VII.6 Calculated Results for Fast Neutrons in Water Using the "Constant a" Theory and the Predicted Values of dE/ds.
<table>
<thead>
<tr>
<th>Liquid Pressure Lbf/ft²</th>
<th>T_{sat}(F₁) Deg F</th>
<th>Vapor Pressure Lbf/ft²</th>
<th>Gas Pressure Lbf/ft²</th>
<th>(dE/ds)_{avg} Mev/cm</th>
<th>ΔE Mev</th>
<th>T_{sup} Deg F</th>
<th>ΔT_{sup} Deg F</th>
</tr>
</thead>
<tbody>
<tr>
<td>2116.2</td>
<td>212.000</td>
<td>27297.3</td>
<td>443.1</td>
<td>2.61(10³)</td>
<td>0.102</td>
<td>377.050</td>
<td>165.050 6.07</td>
</tr>
<tr>
<td>2880.0</td>
<td>227.960</td>
<td>28167.8</td>
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<td>0.101</td>
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<td>151.700 6.07</td>
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<td>267.250</td>
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<td>505.8</td>
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<td>0.097</td>
<td>388.850</td>
<td>121.600 6.07</td>
</tr>
<tr>
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<td>292.710</td>
<td>34567.4</td>
<td>553.7</td>
<td>2.63(10³)</td>
<td>0.093</td>
<td>397.130</td>
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</tr>
<tr>
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<td>600.9</td>
<td>2.64(10³)</td>
<td>0.090</td>
<td>404.730</td>
<td>92.700 6.07</td>
</tr>
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<td>40746.9</td>
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<td>83.940 6.07</td>
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<td>43773.7</td>
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<td>62.640 6.07</td>
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<td>0.076</td>
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<td>46.530 6.07</td>
</tr>
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<td>0.069</td>
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<td>44.660 6.07</td>
</tr>
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<td>0.068</td>
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<td>42.930 6.07</td>
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</table>

Table VII.7 Calculated Results for Fast Neutrons in Water Using the "Constant a" Theory and Values of dE/ds Representing a Positive Uncertainty in the Mean Charge of 20%.
### Table VII.8 Calculated Results for Fast Neutrons in Water Using the "Constant a" Theory

<table>
<thead>
<tr>
<th>Liquid Pressure $P_1$ (Lbf/ft$^2$)</th>
<th>$T_{\text{sat}}(P_1)$ (Deg F)</th>
<th>Vapor Pressure (Lbf/ft$^2$)</th>
<th>Gas Pressure (Lbf/ft$^2$)</th>
<th>$(dE/\text{ds})_{\text{avg}}$ (Mev/cm)</th>
<th>$\Delta E$ (Mev)</th>
<th>$T_{\text{sup}}$ (Deg F)</th>
<th>$\Delta T_{\text{sup}}$ (Deg F)</th>
<th>&quot;a&quot;</th>
</tr>
</thead>
<tbody>
<tr>
<td>2116/2</td>
<td>212.000</td>
<td>37757.5</td>
<td>609.7</td>
<td>$1.46 \times 10^3$</td>
<td>0.037</td>
<td>404.910</td>
<td>192.910</td>
<td>6.07</td>
</tr>
<tr>
<td>2880.0</td>
<td>227.960</td>
<td>38591.8</td>
<td>622.4</td>
<td>$1.46 \times 10^3$</td>
<td>0.036</td>
<td>406.860</td>
<td>178.900</td>
<td>6.07</td>
</tr>
<tr>
<td>5760.0</td>
<td>267.250</td>
<td>41691.4</td>
<td>669.2</td>
<td>$1.46 \times 10^3$</td>
<td>0.035</td>
<td>413.830</td>
<td>146.580</td>
<td>6.07</td>
</tr>
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<td>8640.0</td>
<td>292.710</td>
<td>44752.6</td>
<td>715.5</td>
<td>$1.46 \times 10^3$</td>
<td>0.034</td>
<td>420.330</td>
<td>127.620</td>
<td>6.07</td>
</tr>
<tr>
<td>11520.0</td>
<td>312.030</td>
<td>47775.8</td>
<td>761.3</td>
<td>$1.47 \times 10^3$</td>
<td>0.034</td>
<td>426.420</td>
<td>114.390</td>
<td>6.07</td>
</tr>
<tr>
<td>14400.0</td>
<td>327.810</td>
<td>50767.5</td>
<td>806.1</td>
<td>$1.47 \times 10^3$</td>
<td>0.033</td>
<td>432.160</td>
<td>104.350</td>
<td>6.07</td>
</tr>
<tr>
<td>17280.0</td>
<td>341.250</td>
<td>53723.1</td>
<td>850.9</td>
<td>$1.47 \times 10^3$</td>
<td>0.032</td>
<td>437.580</td>
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<td>$1.48 \times 10^3$</td>
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<td>473.140</td>
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<td>0.027</td>
<td>484.120</td>
<td>55.150</td>
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Table VII.8 Calculated Results for Fast Neutrons in Water Using the "Constant a" Theory and Values of $dE/ds$ Representing a Negative Uncertainty in the Mean Charge of 20%. 

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**Note:** The values of vapor pressure, liquid pressure, and gas pressure are given in Lbf/ft$^2$. The table also includes the calculated values of $(dE/\text{ds})_{\text{avg}}$ in Mev/cm, $\Delta E$ in Mev, and the corresponding values of $T_{\text{sup}}$ and $\Delta T_{\text{sup}}$ in Deg F. The parameter "a" is also provided in the last column.
Figure VII.6 Comparison of the Final Theories for the Threshold Superheat for Fast Neutrons in Water with the Data.
Chapter VIII

Application of the "Closed Energy Balance Method" to Sodium

The "Closed Energy Balance Method" indicates that the theory is completely determinate and can give the superheat threshold as a function of pressure, substance, and type of radiation without any unknown parameters. The theory becomes "closed" when "a" is determinable. Two methods for obtaining "a" were suggested in the previous chapter. One was simply an empirical result for water and is subject at the present time to considerable uncertainty. It would indeed be risky to apply this result to sodium. The other approach is based on a reasonable physical model and appears to be applicable to any situation. Therefore the model approach will be used for the sodium threshold superheat predictions.

VIII.1 Energy Deposition by Heavy Particles in Sodium

The energy deposition by a fission fragment or a primary knock-on sodium atom would again follow the characteristic curve as shown in figure III.1. The fate of the energy which is lost by the particle is a question which must be
considered more closely in the case of sodium.

For the water cases it can be assumed that all the energy transferred to the electrons of the stopping medium is converted to molecular motion in the immediate vicinity of the particle's track. Any attempt by these electrons to transport the energy away from this vicinity by physically moving radially away from the track would produce electric fields due to charge separation. Their departure would therefore be impeded. In sodium the situation is completely different. Metals in general do not possess valance electrons which can be designated to a particular atom. The valance electrons exist in a virtually unbound state and are afforded a great amount of freedom of motion and interaction. This accounts for the high thermal and electrical conductivities of metals as opposed to dielectrics such as water. The effect of this freedom of motion and interaction is to relax in a very short period of time any perturbations which may exist in this electron "cloud" as it is sometimes called. Such a perturbation exists following the passage of a charged particle through the metal. A localized region of highly energetic electrons is created. Through electron-electron interactions this energetic region will very quickly be dissipated without requiring charge separation. Killias (33) indicates that the relaxation time due to electron-electron interactions is a couple orders of magnitude shorter than the relaxation
time due to electron-phonon interactions for the solid state. Seitz (36) develops a relation for the relaxation time for free electrons to transfer energy to the lattice for the solid state situation. Doing the calculation for sodium gives a relaxation time of about $10^{-8}$ sec. This can be compared to the relaxation time for energy loss from an "electron spike" or region of highly excited electrons. This can be estimated to be on the order of $10^{-11}$ sec. Again the energy transfer from the electronic system where it is initially placed by the radiation to the atoms is a relatively slow process. The liquid state would probably not alter the dominance of the relaxation phenomenon by the electron-electron interactions.

The impact of this discussion on radiation induced nucleation in sodium is that the portion of the track where electronic energy transfer is dominant is rendered ineffective for embryo production. Only the end of the track where nuclear elastic collisions are dominant is useful. Therefore, the energy available for the nucleation process will be greatly reduced compared with the energy potential in the electronic portion of the track. Consequently very high superheats must be expected in order for radiation induced nucleation to occur in sodium.
VIII.2 Calculation of Threshold Superheats for Fission Fragments in Sodium

The energy deposition rate for fission fragments in sodium can again be obtained from equations V.1, V.2, V.3, and V.4 with the subscript i referring to sodium. The mean ionization potential was taken as 132 ev from Evans (17). The final equations for the most probable fission fragments are:

\[
\frac{dE}{ds}^L = \rho_i(T_1) \left[ 3.1234 \times 10^4 \ln(0.0286E) + 0.2515 \times 10^4 \ln(8.3603E) \right]
\]

\[
\frac{dE}{ds}^H = \rho_i(T_1) \left[ 3.9479 \times 10^4 \ln(0.0179E) + 0.4742 \times 10^4 \ln(4.005EV) \right]
\]

The first terms in these equations represent the electronic energy transfer and will be neglected due to the above discussion. The second terms are plotted in figure VIII.1. It can be seen that the heavy fragment gives the maximum energy deposition. If the energy of formation is less than 0.6 Mev, the energy deposition rate divided by the density can be taken as constant equal to 10100 Mev cm\(^2\)/gm.

Equation III.14 is the general criterion for the determination of the threshold superheat. For sodium there are no terms of the type \(\Delta E_x\) which need be taken into
Figure VIII.1 Energy Deposition Rates for Fission Fragments in Sodium for the Nuclear Elastic Regime.
account. The equation then becomes:

\[
\frac{4}{3} \pi r^3 \left[ \frac{P^*L}{2} + \rho_v h_{fg} \right] = r^* (dE/ds)_\text{avg}.
\]

\[
\frac{4}{3} \pi r^3 \left[ \sigma/r^* + \rho_v h_{fg} \right] = \rho_l (6.07) (10100) \quad \text{VIII.3}
\]

A simple routine can be set up to calculate the threshold superheat as a function of pressure.

1. Select a value of \( P_1 \)
2. Select an initial guess for the superheat temperature, \( T_1 = T_v \)
3. Calculate \( P, \rho_v, \rho_l, h_{fg} \), and \( \sigma \) from the equations in Appendix A/V
4. Calculate \( r^* \) using equation II.10
5. Substitute into equation VIII.3 and check for satisfaction
6. If VIII.3 is not satisfied, select a new value of \( T_1 \) for the same \( P_1 \) and repeat steps 3, 4, and 5

Sup-routine-C.E.B.M.-F.F.

The results are tabulated in table VIII.1 for pressures from about 1 psia to 150 psia. Higher pressure calculations were not possible because the properties of sodium become uncertain above 2600 °F (See Appendix A). Figure VIII.2 also shows these results.
<table>
<thead>
<tr>
<th>Liquid Pressure Lbf/ft(^2)</th>
<th>(T_{\text{sat}(P_1)}) Deg F</th>
<th>Vapor Pressure Lbf/ft(^2)</th>
<th>Gas Pressure Lbf/ft(^2)</th>
<th>((dE/ds)_{\text{avg}}) Mev/cm</th>
<th>(\Delta E) Mev</th>
<th>(T_{\text{sup}}) Deg F</th>
<th>(\Delta T_{\text{sup}}) Deg F</th>
<th>&quot;a&quot;</th>
</tr>
</thead>
<tbody>
<tr>
<td>144.0</td>
<td>1200.00</td>
<td>28553.5</td>
<td>0.00</td>
<td>6.53(10^3)</td>
<td>0.463</td>
<td>2319.40</td>
<td>1119.40</td>
<td>6.07</td>
</tr>
<tr>
<td>309.0</td>
<td>1300.00</td>
<td>28750.4</td>
<td>0.00</td>
<td>6.52(10^3)</td>
<td>0.462</td>
<td>2321.90</td>
<td>1021.90</td>
<td>6.07</td>
</tr>
<tr>
<td>608.0</td>
<td>1400.00</td>
<td>29107.3</td>
<td>0.00</td>
<td>6.52(10^3)</td>
<td>0.459</td>
<td>2326.40</td>
<td>926.40</td>
<td>6.07</td>
</tr>
<tr>
<td>1116.0</td>
<td>1500.00</td>
<td>29701.2</td>
<td>0.00</td>
<td>6.51(10^3)</td>
<td>0.455</td>
<td>2333.80</td>
<td>833.8</td>
<td>6.07</td>
</tr>
<tr>
<td>1924.0</td>
<td>1600.00</td>
<td>30649.8</td>
<td>0.00</td>
<td>6.49(10^3)</td>
<td>0.448</td>
<td>2345.40</td>
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<td>661.90</td>
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</tr>
<tr>
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<td>1800.00</td>
<td>34036.0</td>
<td>0.00</td>
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<td>0.425</td>
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<td>584.80</td>
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</tr>
<tr>
<td>7320.0</td>
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<td>0.00</td>
<td>6.40(10^3)</td>
<td>0.408</td>
<td>2415.00</td>
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<td>6.07</td>
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<td>10580.0</td>
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<td>40531.9</td>
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<td>6.35(10^3)</td>
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<td>0.00</td>
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<td>0.359</td>
<td>2501.80</td>
<td>401.80</td>
<td>6.07</td>
</tr>
<tr>
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<td>2200.00</td>
<td>53232.0</td>
<td>0.00</td>
<td>6.19(10^3)</td>
<td>0.315</td>
<td>2566.80</td>
<td>366.80</td>
<td>6.07</td>
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</table>

Table VIII.1  Predicted Threshold Superheats for Fission Fragments in Sodium Using the "Constant a" Theory.
Figure VIII.2 Superheat Threshold for Heavy Fission Fragments and Uranium Knock-on in Sodium.
VIII.3 Calculation of Threshold Superheats for Fast Neutrons in Sodium

Again using equations V.1, V.2, V.3, and V.4 with the subscript 1 referring to sodium, the energy deposition rate of sodium knock-on atoms can be obtained. As in the case of the fission fragments the electronic energy deposition can be neglected leaving only the nuclear elastic portion. Equation VIII.4 gives the appropriate relation and figure

\[
\frac{dE}{ds} = \rho_1(T_1) \frac{49.9530}{E} \ln(96.5612E) \quad \text{VIII.4}
\]

VIII.3 shows the \(dE/ds\) curve as a function of energy. If the energy of formation is less than 0.02 Mev, a constant value of \((dE/ds)/\rho_1\) can be used for determining the threshold superheat. The attempted calculations used \((dE/ds)/\rho_1 = 1750\).

It is interesting to note that this same energy deposition curve can be produced by all primary knock-on atoms with energies of 50 kev or greater. Therefore the initial energy of the knock-on sodium atoms is unimportant if greater than 50 kev. Using equations VI.1 and VI.2, the minimum neutron energy capable of producing primary knock-ons of 50 kev in a head-on collision is 0.313 Mev. A very large number of knock-ons of this type could be expected in a fast reactor.
Figure VIII.3 Energy Deposition Rate for Primary Knock-on Sodium Atoms in Sodium.
The calculational procedure as used in the previous section can be used for the fast neutrons or sodium knock-ons. The calculations were attempted but the liquid temperature was consistently above 2600°F and superheat results could not be obtained. This would, however, indicate that the superheat threshold at atmospheric pressure would be greater than 1000°F and perhaps closer to 1500°F.

VIII.4 Minimum Possible Superheat for Radiation Induced Nucleation in Sodium

It is apparent from figures VIII.1 and VIII.3 that the largest energy deposition rate would be obtained for an energetic particle with the greatest possible mass and atomic number. This type of particle could be something like a uranium knock-on atom. Figure VIII.4 shows the energy deposition rate of such a heavy knock-on in sodium based on the same basic equations used for fission fragments. The expression for \(\frac{dE}{ds}\) for this heavy knock-on is given by equation VIII.5. This calculation indicates

\[
\frac{dE}{ds}\,,\,\text{heavy knock-on} = \rho(T) \frac{3.61 \times (10^4)}{E} \ln(1.33E)
\]

that the maximum energy deposition rate divided by the density which can possibly be obtained is about 17,500
Figure VIII.4 Energy Deposition Rate of a Uranium Knock-on in Sodium.
Mev cm$^2$/gm. To provide this $(dE/ds)/\rho_1$ would require a heavy knock-on of about 2 Mev. This amount of energy rules out such things as alpha decay recoils which may have energies on the order of 0.1 Mev. The only conceivable way these energetic heavy knock-ons can be produced is in a "hard" elastic scattering event between a fission fragment and the heavy atom.

The threshold superheats for an energetic uranium knock-on are tabulated in table VIII.2 and are shown in figure VIII.2. These particles do reduce the superheat threshold considerably, but even for this most optimistic case the superheats are in the range of 300 to 400°F.
<table>
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<tr>
<th>Liquid Pressure Lbf/ft²</th>
<th>$T_{\text{sat}}(P_1)$ Deg F</th>
<th>Vapor Pressure Lbf/ft²</th>
<th>Gas Pressure Lbf/ft²</th>
<th>$(dE/ds)_{\text{avg}}$ Mev/cm</th>
<th>$\Delta E$ Mev</th>
<th>$T_{\text{sup}}$ Deg F</th>
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<td>144.0</td>
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<td>1.233</td>
<td>2217.80</td>
<td>717.80</td>
<td>6.07</td>
</tr>
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<td>0.00</td>
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<td>1.203</td>
<td>2233.60</td>
<td>633.60</td>
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<td>0.00</td>
<td>1.12$(10^4)$</td>
<td>1.000</td>
<td>2366.40</td>
<td>366.40</td>
<td>6.07</td>
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<td>0.00</td>
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<td>0.932</td>
<td>2419.10</td>
<td>319.10</td>
<td>6.07</td>
</tr>
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<td>2200.00</td>
<td>43338.9</td>
<td>0.00</td>
<td>1.09$(10^4)$</td>
<td>0.854</td>
<td>2480.20</td>
<td>280.20</td>
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<td>0.744</td>
<td>2553.80</td>
<td>253.80</td>
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</tr>
</tbody>
</table>

Table VIII.2 Predicted Threshold Superheats for 2 Mev Uranium Knock-ons in Sodium Using the "Constant a" Theory.
Chapter IX

Summary, Conclusions, and Recommendations

IX.1 Summary

An attempt has been made in this work to establish a general theory for predicting the conditions under which radiation induced nucleation can occur. The theory is developed in a general form which is applicable to any substance and any type of radiation under any conditions. It must be kept in mind that the theory itself is only as good as the inputs which are fed into it. These inputs are sufficiently general to allow for the peculiarities which may exist in the substance being nucleated and the type of radiation causing the nucleation.

The phenomenon of radiation induced nucleation falls into the category of homogeneous nucleation since it occurs in the bulk of the liquid and is not dependent on surfaces, gas content, dirt particles, etc. With this being the case the conditions for radiation induced nucleation should be obtainable from a homogeneous nucleation treatment. Indeed this is the case as shown in chapter II using a modified Volmer approach. This so called "statistical theory" can take into account the effects of radiation reaction rate,
time period of observation, and system volume and is in
general the correct theory for predicting nucleation condi-
tions. In the absence of radiation this theory predicts
the pure homogeneous nucleation conditions or "foam limit"
as it is sometimes called.

The statistical theory is found to be approximated
very well in nearly all cases except near the foam limit
by a much simpler macroscopic theory called the "energy
balance method." This theory neglects the possibility of
embryo growth through the statistical addition of molecules
and simply demands that the energy available from the radi-
tion be equal to the energy of formation of a critical
embryo. The energy of formation is a function of the liquid
pressure and superheat only for a particular substance and
is determined from the first law of thermodynamics. The
energy available for the formation process depends on the
type of radiation, the track length of the energetic particle
which is effective in forming a single bubble, and the
substance involved.

The peculiarities of the type of radiation and substance
can be handled reasonably well, but the effective track
length renders the theories incomplete. In order to obtain
a closed theory this length must be known. A model based
on the vapor jet instability in a liquid was utilized in this
work. It was found that this effective track length divided
by the critical bubble radius was essentially constant for
all types of radiation, all temperatures and pressures, and all substances. This dimensionless effective track length was found to be 6.07.

The two theories were applied to and compared with two experimental efforts. One was with fission fragments in water and the other was with fast neutrons in water. An experimental apparatus was developed which was capable of generating superheats in water exceeding 100°F and capable of maintaining the superheat for time periods on the order of hours. A 1 cm³ drop of water was suspended and superheated between a heavy and a light oil thereby eliminating all surfaces. The superheat thresholds were obtained by slowly increasing the temperature of the entire system at a particular pressure until boiling was visually observed. The fission fragments were generated directly in the water by adding a small amount of uranium nitrate to the water and exposing the system to neutrons from PuBe neutron sources. The fast neutron data used only the PuBe neutrons thereby producing primary knock-on oxygen atoms in the water drop. These atoms, then, induced the nucleation.

The comparison of the theoretical and experimental results for the two types of radiation indicates fairly good agreement in light of the uncertainties involved. It is difficult to assess the accuracy of the instability model or "Constant a" theory used for determining the effective radiation track length, but it appears to do an adequate
job. The data for both types of radiation can be correlated quite well with an empirical relation for the dimensionless effective track length. However, due to the uncertainties in the data it has limited usefulness.

With the demonstrated adequacy of the energy balance method verses the statistical method for determining threshold superheats and with reasonable confidence in the instability model for determining the effective track length, the threshold superheats for sodium were predicted. In these calculations the energy transferred to the electronic system of the sodium was assumed to be dissipated throughout the electronic system instead of being transferred to the sodium molecules directly in the vicinity of the particle's track. The thermal spike was assumed to be formed only at the end of the particle's track where nuclear elastic collisions are dominant. Results are presented for the heavy fission fragments and for the uranium knock-on atom which is the most optimistic particle from the standpoint of requiring the lowest threshold superheat.

The application of the "closed energy balance method" to sodium under the assumption that the energy deposited in the electronic system is ineffective in the formation process gives threshold superheats for fast neutrons which are considerably above the range of usefulness from the safety standpoint in a liquid metal cooled fast reactor. Fission fragments and heavy knock-on atoms of about 2 Mev appear to be the only particles which could be effective in pro-
viding a reasonable upper limit on superheat in such a reactor. Even for these most optimistic particles the superheat limit established through radiation induced nucleation is too high to have a significant impact on the LMFBR void problem. This phenomenon does not appear to offer a basis for relaxing design considerations by providing a much reduced absolute ceiling on the maximum possible superheat. It might be justifiably argued, however, that there is no basis for placing the maximum possible superheat for design purposes at a value greater than 300 to 400 °F depending on the system pressure.

IX.2 Conclusions

1. The homogeneous theory of Volmer (6) can be modified to take into account embryos produced by nuclear radiation as in chapter II.

2. The "energy balance method" is a special case of the more general homogeneous or statistical theory of chapter II for the case of very small embryo inputs from radiation.

3. The "energy balance method" will give very good results for all realistic system except very near the "foam limit." Calculational procedures for both approaches are given in section V.3 and section V.4.
4. The effective track of the radiation which is effective in the formation of a single bubble appears to be given quite adequately by the "Constant a" theory as outlined in section VII.5. This theory predicts a constant value of the dimensionless length "a", which is defined as the actual effective track length divided by the critical bubble radius, of 6.07. It appears that "a" has the same value for all substances, types of radiation, and system conditions.

5. The theoretical results of the "closed energy balance method" where "closed" indicates a completely determinate theory are reasonably verified by experimental results for fission fragments and fast neutrons in water at low pressure. These comparisons are shown in figures VII.5 and VII.6.

6. In applying this theory to sodium the energy available for bubble formation is diminished by the ineffective-ness of the energy deposited in the sodium electronic system in forming a thermal spike and vapor column.

7. The superheat thresholds predicted by the "closed energy balance method" for fission fragments in sodium, equation VIII.3, are on the order of 400 °F at about 100 psia.

8. The lowest possible superheat threshold for sodium is brought about by a heavy knock-on atom such as uranium of about 2 Mev as shown in figure VIII.2. These lowest
possible superheats are around 300 °F at about 100 psia.

9. Fast neutrons in sodium require threshold superheats in the range of 1000 to 1500 °F in the low pressure range.

10. In general it does not appear that nuclear radiation induced nucleation can be expected to have a significant impact on the sodium void problem in the sodium cooled fast breeder reactor or fast reactor safety as a whole.

IX.3 Recommendations

One area in which some continued effort should be made is in reducing the experimental uncertainties in the data presented in this work. This step could lead to a more definitive determination of the correctness of the instability model for the effective track length. In this same regard it would be helpful if the uncertainty in the mean charge of the energetic particle could be reduced.

The whole area concerning the fate of energy transferred to the electronic system of a good conductor should be investigated. The ineffectiveness of the energy transferred to the electrons of the sodium constitutes a factor of 2 increase in the superheat threshold for fission fragments.
If there is any chance that this energy is effective, then some of the conclusions regarding LMFBR design considerations could be altered.

In the final analysis it will be necessary to attempt to experimentally measure the threshold superheat for sodium. Some thought should be given as to how this might best be done if indeed it can be done.
APPENDIX A

Functional Forms of Material Properties

The iterative nature of the calculations performed in this work makes the computer a very desirable tool to use. In order to adapt the theory to the computer it is helpful to have the required physical and thermodynamic properties in functional form. The relationships used in this work for both water and sodium will be presented along with comparative plots in the cases where empirical relations were used for existing data.

A.1 Relations for Water

The properties required in the water calculations are $P_v(T)$, $\rho_v(T)$, $\rho_l(T)$, $h_{fg}(T)$, and $\sigma(T)$. For purposes of this work it is desirable to have these relations for the temperature range of 200°F to 450°F.

The vapor pressure relationship from Keenan (32) is:

$$\log_{10}\left[\frac{P_c}{P_v}\right] = \frac{1}{2.3} \ln e\left[\frac{P_c}{P_v}\right] = \frac{K}{T} \left[\frac{a+bx+cx^3+dx^4}{1+dx}\right]$$

A.1
Where: 
\( P_v \) = vapor pressure in atm

\( P_c \) = critical pressure = 218.167 atm.

\( T \) = temperature in °K

\( x = (T_c - T) \)

\( T_c \) = critical temperature = 647.27°K

\( a = 3.3463130 \)

\( b = 4.14113(10^{-2}) \)

\( c = 7.515484(10^{-9}) \)

\( d = 1.3794481(10^{-2}) \)

\( e = 6.56444(10^{-11}) \)

This equation is valid for 50°C ≤ T ≤ 347°C.

The surface tension relationship was obtained by fitting a straight line through the data presented in Tipton (33) using the "least squares" methods. The resulting equation is applicable from 200°F to 450°F.

\[ \sigma(T) = -8.198(10^{-6})T(°F) + 0.005738 \text{ Lbf/ft.} \]  

A comparison of this equation and the data of Tipton (33) is shown in figure A.1.

The liquid density was also obtained from Keenan (32). This relationship is valid for saturated liquid from 32°F to 680°F. The fact that superheated liquid is being approximated by saturated liquid at the same temperature should not introduce significant error.
\[
\rho_1(T) = \frac{1}{v_1} = \left[ \frac{v_c + ax^{1/3} + bx + cx^4}{1 + dx^{1/3} + ex} \right]^{-1}
\]

Where:
- \( v_1 \) = specific volume \( \text{cm}^3/\text{gm} \)
- \( x = (T_c - T) \)
- \( T_c \) = critical temperature 374.11\(^\circ\)C
- \( T \) = temperature \( ^\circ\)C
- \( v_c = 3.1975 \text{ cm}^3/\text{gm} \)
- \( a = -0.3151548 \)
- \( b = -1203374 \times 10^{-3} \)
- \( c = 7.48908 \times 10^{-13} \)
- \( d = 0.1342489 \)
- \( e = -3946263 \times 10^{-3} \)

The density of the vapor inside the embryos may be calculated using equation 13 of Keenan (32).

\[
\rho_v(T) = \frac{1}{v_v} = \left[ \frac{4.55504T(\text{OK})}{P} + B \right]^{-1}
\]

Where:
- \( B = B_0 + B_0^2g_1(3/1)P/T + B_0^4g_2(3/1)P^3/T^3 - B_0^{13}g_3(3/1)P^{12}/T^{12} \)
- \( \rho_v \) = density in \( \text{gm/cm}^3 \)
- \( T \) = temperature in \( ^\circ\)K = 273.16 + \( T(\text{\circ}C) \)
$P = \text{pressure in atmospheres}$

$B_0 = 1.89 - 2641.62(1/T)10^{80870/T^2}$

$g_1(1/T) = 82.546/T - 1.6246(10^5)/T^2$

$g_2(1/T) = 0.21828 - 1.2697(10^5)/T^2$

$g_3(1/T) = 3.635(10^{-4}) - 6.768(10^{64}/T^{24}$

The pressure appearing in this expression is the vapor pressure at $T$ and can be obtained from equation A.1.

An expression for $h_{fg}(T)$ was obtained by fitting the tabulated data of Keenan (32) with a second order polynomial for the temperature range from 200°F to 500°F.

$h_{fg}(T) = 1064.6 - 0.270T - 8.15(10^{-4})T^2 \text{ BTU/LBm} \quad \text{A.5}$

$T$ in this equation is in degrees F. A comparison of the tabulated values from the steam tables of Keenan with this equation is shown in figure A.2.

The remaining input to be considered is the condensation co-efficient, $\bar{X}$. Rohsenow (34) indicates that values of the condensation coefficient are very hard to predict. For very pure fluids the value is probably close to unity, but the maximum measured value for water is 0.04. On this basis an order of magnitude value of 0.01 was chosen for use in computations.
Figure A.1 Comparison of Tabulated Values of the Surface Tension for Water with the Empirical Expression Used in this Work.
Figure A.2 Comparison of Tabulated Values of the Heat of Vaporization of Water with the Empirical Expression Used in this Work.
A.2 Relations for Sodium

The same properties are required for sodium as for water. All of these properties were taken from Golden and Tokar (35). The relations presented are not assumed to be valid above 2600°F and the sodium calculations were terminated at this temperature.

The expression used for the vapor pressure is equation 3.1 of reference (35) and is considered valid for the temperature range 1624°F to 2539°F.

\[ \log_{10} P_v = 6.83770 - \frac{9980.94}{T} - 0.61344 \log_{10} T \]  

A.6

In modified form this equation becomes:

\[ P_v(T) = \exp\left[15.7267 - \frac{22956.16}{T} - 0.61344 \ln T\right] \]  

A.7

Where:  
\( P_v \) = vapor pressure in atmospheres  
\( T \) = temperature in °R

The surface tension is given by equation 7.9a of reference (35). This expression is assumed to be fairly good up to 2500°F.

\[ \sigma(T) = 0.01429 - 3.81(10^6)T \]  

A.8
Where:  \( \sigma \) = surface tension in Lbf/ft
\[ T = \text{temperature in } ^\circ F \]

The liquid density is given by equation 2.1 of reference (35) for the temperature range from 208\(^\circ\)F to 2500\(^\circ\)F.

\[
\rho_l(T) = 59.566 - 7.9504 \times 10^{-3} T - 0.2872 \times 10^{-6} T^2 + 0.06035 \times 10^{-9} T^3
\]  \( A.9 \)

Where: \( \rho_l \) is the liquid density in Lbm/ft\(^3\)
\( T \) is the temperature in degrees F

This equation is plotted in figure A.3 along with the values from reference (35).

The vapor density expression was obtained by fitting a cubic equation through the vapor specific volume values given by reference (35). The equation is:

\[
\rho_v(T) = \left[ 889.263 - 1.06672 T + 0.432326 \times 10^{-3} T^2 - 0.589483 T^3 \right]^{-1}
\]  \( A.10 \)

Where: \( \rho_v \) is the vapor density in Lbm/ft\(^3\)
\( T \) is the temperature in degrees F

This equation is plotted in figure A.3 along with the values from reference (35).

The heat of vaporization was obtained using a linear fit of the values presented in reference (35). Over the
temperature range of interest here, 1900°F to 2500°F, the linear equation representation is good to less than 1%.

\[ h_{fg}(T) = 2115 - 0.270T \]  

Where: \( h_{fg} \) is the heat of vaporization in BTU/Lbm

\( T \) is the temperature in degrees F
Figure A.3 Comparison of Empirical Representation of Sodium Vapor Density with Tabulated Values.
Figure A.4 Comparison of the Empirical Expression for $h_{fg}$ with Tabulated Values of Golden (35).
Some Considerations of the Interaction of Heavy Charged Particles With Matter

B.1 Relative Energy Deposition Rates of Primary Oxygen Knock-ons and Hydrogen Knock-ons

In treating the fast neutron induced nucleation in Chapter VI it was assumed that the primary knock-on oxygen atoms were the important energy depositors in the process. The energy deposition rates of both the hydrogen and oxygen knock-on should be investigated to substantiate this assumption.

The energy deposition rate for the oxygen knock-on is given by equation VI.4 and is shown plotted as a function of energy in figure B.1. It is assumed that the maximum energy knock-ons from 10.5 Mev neutrons are available, i.e., 2.31 Mev knock-ons. For the same energy neutrons hydrogen knock-on of 10.5 Mev can be produced. Their energy deposition rate as a function of energy can be calculated in the high energy range with an expression like equation 2.1 of Evans (17), Chapter 22. This
\[ \frac{dE}{ds} = \frac{4\pi e^4}{m_0 v^2} N Z \left[ \ln \frac{2m_0 v^2}{I} - \ln(1-\beta^2) - \beta^2 \right] \quad B.1 \]

equation reduces to the following for protons in water neglecting relativistic effects. The reduction in the

\[ \frac{dE}{ds} = \frac{79.5}{E} \ln(31.9E) \quad B.2 \]

mean change in the proton can be factored in using figure 1.1 of Evans (17), Chapter 22. The result for the proton is also shown in figure B.1. The oxygen knock-on at 2.31 Mev is on the low energy side of the Bragg peak while the maximum energy deposition for the proton occurs at the Bragg peak. It is apparent from this figure that the oxygen knock-on should give the greater concentration of deposited energy and should therefore control the fast neutron induced nucleation phenomenon.

B.2 Possible Loss of Energy From the Radiation Track Due to Delta Electrons

The concern regarding delta electrons is that these electrons may be sufficiently energetic to transport energy radially away from the critical region in which nucleation
must occur. This effect could only be important in the case of water since the electronic energy transfer portion of the track is unimportant in sodium. Also if the effect is important it will affect the fast neutron case the most since the critical region is smaller.

By the critical region is meant the cylindrical region in the liquid, which is concentric with the path of the particle, into which the deposited energy must be contained if it is to be effective in the nucleation process. This cylinder is essentially a cylinder of liquid which contains the final mass in the vapor cylinder which results. The radius of this critical region is given by equation B.4

\[
\pi (r_c)_v^2 \rho_v L = \pi (r_c)_1^2 \rho_1 L \quad \text{B.3}
\]

\[
(r_c)_1 = (r_c)_v \sqrt{\rho_v / \rho_1} \quad \text{B.4}
\]

in terms of the radius of the vapor cylinder. The radius of the vapor cylinder can in turn be related to the initial bubble radius by:

\[
(Volume)_\text{sphere} = (Volume)_\text{cylinder}
\]

\[
\frac{4}{3} \pi r^3 = \pi (r_c)_v^2 a_r^* 
\]
\( (r_c)_v \approx 0.47 r^* \) for \( a = 6.07 \) \( B.5 \)

Therefore, the critical region in the liquid in which the energy must be deposited is defined by the radius given in equation \( B.6 \). For the fast neutron cases considered in this work the temperature might be around 450°F and \( r^* \)

\( (r_c)_1 = 0.47 r^* \sqrt{\rho_v/\rho_l} \) \( B.6 \)

would be about \( 4(10^{-6}) \) cm giving \( (r_c)_1 \) as \( 0.26(10^{-6}) \) cm.

The maximum energy which can be transferred to an atomic electron is \( 2M_e V^2 \) if the mass of the particle is much greater than the mass of the electron, \( m_e \). For a 2.3 Mev oxygen knock-on this maximum energy is \( 3.15(10^{-4}) \) Mev. The range of this maximum energy delta electron by the "Thomson-Whiddington law" from Evans (17) is approximately \( 0.2(10^{-6}) \) cm. Then if this maximum energy delta electron were traveling radial outward from the axis of the track it would still be in the critical zone when it stopped and would therefore deposit all its energy within the zone.

In general the energy carry-off by delta electrons can be neglected except perhaps at very high pressures where \( (r_c) \) becomes very small. Even in the high pressure case, however, the probability of obtaining these high energy delta electrons is such that the overall effect may still be negligible.
Figure B.1 Comparison of Energy Deposition Rates for Hydrogen and Oxygen Knock-ons from 10.5 Mev Neutrons.
Appendix C

Energy of Formation of a Nucleus from the First Law of Thermodynamics

Consider again figure II.2 for a system of mass \( m \) with and without a nucleus. The First Law for this change of state is:

\[
U_2 - U_1 = Q_2 - W_2 \tag{C.1}
\]

\[
(m - m_v)u_1 + m_vu_v - m_1u_1 = E^*_f - P_1m_v(v_v - v_1) - \sigma A^* 
\]

\[
u_v - u_1 = \frac{E^*_f}{m_v} - P_1v_v + P_vv_1 - \sigma A^*/m_v
\]

\[
(u_v + P_vv_v) - (u_1 + P_1v_1) = \frac{E^*_f}{m_v} - P_1v_v + P_vv_v - \sigma A^*/m_v
\]

\[
E^*_f = m_v(h_v - h_1) + \sigma A^* - m_vv_v(P_v - P_1) \tag{C.2}
\]

Substituting \( A^* = 4\pi r^2 \), \( m_v = \rho_v(4/3)\pi r^3 \), \( v_v = 1/\rho_v \), and \( (P_v - P_1) = 2\sigma/r^* \) into equation C.2 gives:

\[
E^*_f = (4/3)\pi \left[ \rho_v(h_v - h_1) + \frac{1}{2}(P_v - P_1) \right] r^3 \tag{C.3}
\]

If the change in the enthalpy is taken as the heat of vapor-
ization which is a very good assumption, equation C.3 becomes equivalent to equation III.6 of chapter III.
## Appendix D

### Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>Surface area of an embryo</td>
</tr>
<tr>
<td>D</td>
<td>Thermal diffusivity</td>
</tr>
<tr>
<td>E</td>
<td>Energy of the energetic particle passing through the stopping medium</td>
</tr>
<tr>
<td>E_A</td>
<td>Energy available from the radiation track for nucleation</td>
</tr>
<tr>
<td>E_n</td>
<td>Energy of a neutron before scattering</td>
</tr>
<tr>
<td>E_n0</td>
<td>Maximum neutron energy in a neutron spectrum</td>
</tr>
<tr>
<td>F</td>
<td>Helmholtz free energy</td>
</tr>
<tr>
<td>F_R</td>
<td>Qualitative free energy</td>
</tr>
<tr>
<td>G(H_2)</td>
<td>Yield of hydrogen gas molecules in radiolysis of water</td>
</tr>
<tr>
<td>I_i</td>
<td>Mean ionization potential of the i-th component in the stopping medium</td>
</tr>
<tr>
<td>J</td>
<td>Net rate of reaction per unit volume</td>
</tr>
<tr>
<td>J^-</td>
<td>Net reaction rate toward smaller sizes per unit volume</td>
</tr>
<tr>
<td>K.E.</td>
<td>Kinetic energy</td>
</tr>
<tr>
<td>L</td>
<td>Effective length of the radiation track involved in the formation of a single embryo</td>
</tr>
<tr>
<td>M</td>
<td>Molecular weight</td>
</tr>
<tr>
<td>M_1</td>
<td>Mass of the energetic particle</td>
</tr>
</tbody>
</table>
\(M_i\) Mass of the \(i^{th}\) atom species
\(M_N\) Mass of a nucleus
\(M_n\) Mass of a neutron
\(N\) Molecule density
\(N_{Av}\) Avagadro's number
\(N_i\) Density of embryos containing \(i\) molecules each
\(P\) Pressure
\(PR\) Production rate of primary knock-on atoms
\(Q\) Heat of reaction
\(R\) Universal gas constant
\(RRR\) Radiation reaction rate per unit volume
\(R(n_R)\) Production rate of embryos from the radiation tracks per unit volume
\(R(\rightarrow)\) Reaction rate toward larger embryo sizes per unit volume
\(R(\leftarrow)\) Reaction rate toward smaller embryo sizes per unit volume
\(S\) Entropy
\(\mathcal{S}\) Total number of embryos produced in the system from the radiation tracks in a time \(\tau\)
\(T\) Temperature
\(J\) Energy of the primary knock-on atom
\(T_m\) Mean temperature of the water drop used in the experiments
\(T_p\) Temperature measured by the permanent probe
\(T_s\) Temperature of the surface of the drop used in the experiments
\(TR\) Temperature ramp of experimental apparatus
\(\Delta T\) Superheat - also - \(\Delta T_{sup}\)
\[ \Delta T_0 \] Absolute superheat threshold for fast neutrons induced nucleation

\[ \delta T \] Desired uncertainty limit in the threshold superheat determination

\[ U \] Internal energy

\[ V \] Volume

\[ V \] Velocity of the energetic particle passing through the stopping medium

\[ V_{nf} \] Velocity of the nucleus after scattering a neutron

\[ V_{nf} \] Velocity of a neutron after scattering

\[ V_{n_1} \] Velocity of the neutron before scattering

\[ V_s \] System volume

\[ W \] Work of formation of an embryo

\[ W(l \rightarrow v)(1) \] Flux of molecules from the liquid to the vapor embryo of size \( i \) per unit time and area

\[ W(v \rightarrow l)(1) \] Flux of molecules from the vapor embryo of size \( i \) to the liquid per unit time and area

\[ X(n_R) \] Number of embryos of size \( n_R \) formed in each radiation track

\[ Z \] Gibbs free energy

\[ Z_1 \] Atomic number of the energetic particle

\[ Z_i \] Atomic number of the \( i \)th atom species in the stopping medium

\[ (Z_1)_{\text{eff}} \] Effective mean charge of the energetic particle

\[ a \] Dimensionless effective track length = \( L/r^* \)

\[ a_H \] Radius of the first Bohr orbit for the hydrogen atom

\[ a_{1i}^{\text{scr}} \] Maximum impact parameter for the \( i \)th atom species in the stopping medium

\[ b \] Ratio of the pressure difference across the embryo interface to the vapor pressure
c  Velocity of light

\(c_s\)  Concentration of uranium nitrate in the water used in the experiments with fission fragments

dc  Diameter of the vapor cylinder

e  Magnitude of the charge on an electron

f  Helmholtz free energy per unit mass

f  Qualitative free energy per unit mass

h  Enthalpy per unit mass

h  Heat transfer coefficient

h  Plank's constant

\(\hbar\)  Plank's constant divided by \(2\pi\)

hf\(_{fg}\)  Heat of vaporization per unit mass

k  Boltzmann's constant

m  Mass of a molecule

m  Mass of a particular system

m\(_0\)  Mass of an electron

nR  Number of molecules in the embryos produced from radiation

\(\rho\)  Probability function

q  Rate of heat transfer

r  Radius of a spherical embryo

rc  Radius of a vapor cylinder

s  Entropy per unit mass

s  Distance along the radiation track from the starting point of the track

t  time

u  Internal energy per unit mass

v  Volume per unit mass or specific volume
\( \bar{v} \) Mean molecular speed

\( w_g \) Mass of gas in an embryo

\( x(i) \) Ratio of the radius of an embryo of size \( i \) to the critical radius

\( z \) Gibbs free energy per unit mass

**Greek**

\( \alpha \) Original phase of a substance

\( \alpha_i \) A single molecule of the original phase

\( \bar{\alpha} \) Condensation coefficient

\( \beta \) New phase of the substance

\( \beta \) Velocity of the energetic particle divided by the speed of light

\( \beta_i \) embryo of the new phase of \( i \) molecules

\( \Theta \) Scattering angle in the center-of-mass frame of reference

\( \lambda \) Heat of vaporization per molecule

\( \lambda \) Wavelength of surface disturbances on a vapor cylinder in a liquid

\( \mu \) Chemical potential

\( \mu_i \) Chemical potential of the \( i^{th} \) component of a substance

\( \nu_i \) Number of atoms of the \( i^{th} \) species per molecule

\( \rho \) Density

\( \sigma \) Surface tension

\( \bar{\sigma} \) Average fission cross section

\( \sigma' \) Scattering cross section

\( \tau \) Observation time interval in which nucleation must occur
\( \Phi \)  
Scattering angle in the laboratory frame of reference

\( \phi \)  
Total neutron flux - also - \( \phi_T \)

\( \phi \)  
Average chemical potential per molecule

\( \phi_n \)  
Normalized neutron flux

\( \nu_i \)  
Ratio of the forward to reverse reaction probabilities for producing an embryo of \( i \) molecules

\( \Omega \)  
The thermodynamic potential for doing work at constant temperature and chemical potential

\( \Omega' \)  
Solid angle into which the neutron scatters

**Subscripts**

1  
liquid

v  
vapor

s1  
system at state 1

s2  
system at state 2

e1  
environment at state 1

e2  
environment at state 2

m  
number of molecules in the smallest embryo

U  
Uranium

d  
Water drop

g  
Gas

c  
Vapor cylinder

**Superscripts**

*  
Critical conditions

L  
Light fission fragment

H  
Heavy fission fragment
Appendix E

References


12. Trefethen, L., "Nucleation at a Liquid-Liquid Inter-


Biographical Note

The author was born September 9, 1942 in Zanesville, Ohio. After graduation from Zanesville High School in 1960 he entered the University of Cincinnati and graduated in 1965 with the degree of B.S.M.E. In the summer of 1965 he married the former Nancy G. Taylor of Zanesville, Ohio and moved to Cambridge, Massachusetts. He entered the Nuclear Engineering Department at M.I.T. in 1965 and completed the requirements for the degree of Doctor of Philosophy in March 1970.

While at M.I.T. the author held a National Science Foundation Traineeship, Atomic Energy Commission Special Fellowship in Nuclear Science and Engineering, and M.I.T. Assistantships. He is a member of Tau Beta Pi and Pi Tau Sigma honorary societies and a student member of the American Nuclear Society.