RADIATION INDUCED NUCLEATION
IN
WATER AND ORGANIC LIQUIDS

by

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(1970)

Submitted in Partial Fulfillment of

the Requirements for the Degree of

Master of Science

at the

Massachusetts Institute of Technology

March 1972

Signature of Author _____

Certified by _____
Thesis Supervisor, Date

Accepted by _____
Chairman, Departmental Committee
on Graduate Students

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Submitted to the Department of Nuclear Engineering on March 24, 1972 in partial fulfillment of the requirements for the degree of Master of Science.

Abstract

Experimental data for the threshold superheat was obtained for the case of fission fragments in water and in propylene glycol. Analytical results, using Bell's (5) energy balance method, for the threshold superheat values were obtained for both fission fragments and fast neutrons in water, propylene glycol, benzene, acetone, and nitromethane.

Experimental and analytical results were compared with data obtained by other investigators. (3),(4),(5).

A detailed discussion of the liquid suspension method used in this work is given, along with a criteria for selecting a test fluid and its supporting fluids.

Physical properties vs. temperature plots for a number of possible test fluids are also given.

Thesis Supervisor — Neil E. Todreas
Title — Associate Professor

Acknowledgements

I wish to express my sincere gratitude to my thesis supervisor, Professor Neil E. Todreas, for his advice and assistance throughout this study. I would also like to express my appreciation to Dr. Norman C. Rasmussen for being kind enough to be my thesis reader.

Special thanks are also due to Dr. C.R.Bell for his helpful advice. I would like to take this opportunity to thank all members of the M.I.T. reactor machine shop and radiation protection office for their technical advice and assistance.

Last but not least, I would like to thank all those friends in the Nuclear Engineering Department and in the Chemistry Department who had in one way or another assisted me in the present work.

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Chapter 1

Introduction

1.1 Radiation Induced Nucleation

The term radiation-induced nucleation refers to the phenomena of nucleation (the initial formation of bubbles) of vapor or gas bubbles in liquids due to localized energy deposition or localized ionization along the track of an energized particle.

Homogeneous nucleation is normally not considered an important part of the boiling process in engineering components, since nucleation due to the presence of surface cavities, suspended particles, or non-condensible gas bubbles is the dominant mechanism at small liquid superheats. Pure homogeneous nucleation only becomes important at high superheats.

This phenomena of radiation-induced nucleation of bubbles in a superheated liquid was first reported by D.A. Glaser (1), who was searching for an instrument similar to a cloud-chamber-like detector, which would aid in the study of high energy nuclear events. The cloud chamber utilizes the instability of supercooled vapors against droplet formation, so it seemed promising to attempt to devise an instrument which could take advantage of superheated liquids against bubble formation. Glaser

made an experimental test with superheated diethyl ether. In the absence of any radioactive source a superheated state was able to be maintained for a few seconds. But when a radioactive source was brought near the superheated liquid, it immediately erupted into vapor.

This series of experiments showed that bubble chambers could be a practical device for the study of high energy nuclear events, and led to the highly sophisticated bubble chamber technology which exists today.

Since the existence of the radiation-induced nucleation phenomena was thus demonstrated, the question soon arose of whether or not this would have any affect on nuclear reactor coolants or moderators. Recently, it was desired to know whether this phenomena would have any affect on the sodium void problem in sodium cooled fast reactors. Liquid sodium can attain very high superheats on engineering surfaces. Nucleation of this highly superheated sodium will lead to violent flashing, resulting in the sudden ejection of sodium from one or more coolant channels. The rate of sodium ejection is related to the amount of superheat attained. Since a reactor is an intense source of radiation, the possibility that this radiation, through radiation-induced nucleation, might place an upper limit on the superheat attainable in reactor coolants was a factor worth studying.

1.2 Experimental Methods

There are presently two experimental methods employed to study the radiation-induced nucleation phenomena. The major difference between these two methods is in the manner in which the superheated state of the liquid is attained.

The first, and up to now the most widely used method, is the so-called expansion method. The second is a constant pressure type method. Both of these methods will be reviewed, emphasizing both their advantages and disadvantages.

1.2a Expansion Method

The expansion method is similar to the bubble chamber method of operation. The test liquid is heated to a temperature considerably above its normal boiling point, but is kept at a pressure which still maintains it as a stable liquid.(In other words, the test liquid is kept under a pressure higher than the saturated vapor pressure corresponding to the temperature to which the liquid is raised.) The expansion process then occurs. (Expansion is the sudden lowering of the pressure to a predetermined pressure at which the test liquid is superheated.) The pressure is thus reduced from an initial value, at which the liquid is subcooled, to a lower value,where the test liquid is superheated. This method has been used by Becker(2), Deitrich (3), and El-Nagdy (4).

1.2b Constant Pressure Method

This method, devised by Bell (5), attains the superheated state of the test fluid by maintaining a constant pressure and gradually heating the test liquid. Nucleation from the surface of the test liquid does not occur since the test liquid is not in contact with a solid surface. This is accomplished by suspending the test liquid in the form of a bubble (approximately one-half inch in diameter) between two other liquids, a supporting fluid and a cover fluid. A detailed description of this experimental technique will be given in a later section of this work.

1.2c Advantages and Disadvantages

A major disadvantages of the expansion method is the short duration (a few seconds) of the superheated state of the test liquid. Any effect of radiation on the liquid must therefore be observable during this short period of superheat..

If a reactor (as in Deitrich (3)) or an accelerator (as in El-Nagdy (4)) is used as the source of radiation, then a further complication, that of synchronizing the radiation burst with the expansion in the test chamber, is encountered.

Another disadvantage of the expansion method is that of surface cavities. These will cause boiling on surfaces of the chamber well before the radiation - induced nucleation phenomena can appear. (This problem

has been solved to some degree by El-Nagdy (4), by cooling the test chamber walls just prior to and during the expansion process.)

The constant pressure heating method can maintain a high degree of superheat for an indefinite period of time in the absence of radiation since the technique of suspending the test bubble eliminates surface effects.

The one main disadvantage of this method is that the requirements of this three-liquid configuration are such that it is very difficult to find compatible supporting and cover fluids for many test fluids of interest. The expansion method on the other hand has no such liquid compatibility requirement, and can be used for any liquid desired. A detailed discussion of these compatibility requirements will be given later.

1.3 Objectives of Present Work

The constant pressure heating method was begun at M.I.T. by C.R. Bell (5) who formulated a theory and constructed the experimental apparatus. Some preliminary experimental work with water as the test fluid was also done by Bell (5). This work was continued by Tso (6), who extended the experimental work with water, and performed preliminary analytical work with benzene.

The present work included experimental work with water and propylene glycol, and analytical work (utilizing

Bell's method) with water, propylene glycol, benzene, acetone, and nitromethane. Modifications were also made to the experimental apparatus.

Chapter 2

Theory

2.1 General Introduction

Two theories have been proposed to explain the formation of bubbles by radiation. The first theory, the "Electrostatic theory", was proposed by Glaser (7), and the second theory, the "thermal spike" theory, was proposed by Seitz (8).

Actually there are only two ways in which energy can be extracted from ions to produce bubbles: either the energy of the electric field due to the charges of the ions is converted into the work of formation of bubbles, or the recombination energy of the ions is given off as heat, which produces a so called "thermal spike", and finally bubbles.

The thermal spike theory has generally been accepted as the correct theory, but a short discussion on the electrostatic theory will be presented, since it also did a reasonable job of predicting operating conditions for a number of bubble chambers.

This thesis will use the theoretical approach of Bell (5), which is based on the thermal spike theory.

2.2 The Electrostatic Theory

In order to predict the operating conditions for

a bubble chamber, a theory was needed to quantitatively describe the conditions under which ionizing events could nucleate bubbles in a superheated liquid. Glaser(7) then developed his electrostatic theory.

The electrostatic theory was a microscopic model of an ionization-triggered bubble nucleation mechanism. It was assumed that an ionizing particle produces clusters of electric charges of like sign within a liquid, and that the mutual electrostatic repulsion of these charges could then fracture a superheated liquid to produce bubble nuclei large enough to grow to visible size, using the thermal energy from the liquid. An approximate treatment of the effect of electrostatic forces on bubble nucleation was made, which assumed a number "N" (it was known that a number of charges were required for this mechanism to work) of electronic charges to be distributed uniformly over the surface of a potential bubble nucleus. This treatment formulated an "electrostatic pressure effect", which predicted that a bubble carrying "N" like charges would grow if the pressure on the liquid were less than the saturated vapor pressure at the ambient temperature by a certain amount. This difference, $\Delta P = P_\infty(T) - P$, where: P is the pressure on the liquid and $P_\infty(T)$ is the saturated vapor pressure over a flat liquid -vapor interface, is given by:

$$\Delta P > \frac{3}{2}\left(\frac{4\pi}{N^2 e^2}\right)^{1/3}\left[\sigma(T)\right]^{4/3}\left[\mathcal{E}(T)\right]^{1/3} \qquad (2.1)$$

where:

e = electronic charge

$\sigma(T)$ = surface tension of the liquid at temperature T

$\mathcal{E}(T)$ = dielectric constant of the liquid at temperature T

With N taken as 6, this formula had fairly good success in predicting the operating conditions of a number of liquids, such as hydrogen, deuterium, and helium.

Even though the theory met with some initial success, serious objections arose. One of these considered the fact that in order to rupture the superheated liquid, the N charges must be deposited within a sphere of radius "r", where r is given by:

$$r = \frac{2\sigma(T)}{P_\infty(T) - P} = \frac{2}{3}\left(\frac{2N^2e^2}{\pi}\right)^{1/3}\left[\sigma(T)\right]^{-1/3}\left[\mathcal{E}(T)\right]^{-1/3} \quad (2.2)$$

For typical operating conditions for certain liquids, such as some organic liquids, this gives a radius of the order of 10^{-6} cm.. This requires that an ionizing particle must leave a cluster of six charges, of the same sign, in this small volume. It was obvious that this does not happen often enough in primary ionizations to account for the observed number of bubbles.

A contradiction with the theory was also found when alpha particles were used as the ionizing agent. To agree

with this theory, the alpha particle was required to deposit 900 charges, of the same sign, in a region 2×10^{-6}cm. in diameter. This is greater than the maximum ionization attained by an alpha particle during the slowing down process, even granting the possibility of the required charge separation. The total energy loss of the alpha particle, however, was sufficient to explain the observed effects, so another model for the microscopic mechanism was sought. (For a more detailed description of the electrostatic theory and its disadvantages, references (4), (9), and (10) are recommended.)

2.3 Thermal Theory

The thermal theory suggests that local heating by ions and δ -rays (knocked-on electrons) could produce bubbles along the track of ionized particles.

It was assumed that the primary mechanism for the deposition of this local thermal energy occurred through the δ -rays formed by the primary ionizing particle. These knock-on electrons would lose energy by collisions with molecules. The energy of the δ-ray is thus rapidly converted to kinetic energy of the molecules of the medium. The primary ionizing particle thus produces along its path a series of "thermal spikes", which then act as nuclei for bubble formation.

Seitz (8) suggested that this thermal energy could also be obtained from the heat energy released

in ion recombination at the end of δ -rays, rather than just coming from molecular excitation.

The basic assumption of this thermal spike model is that the minimum energy required to form a bubble of critical size is provided from the thermal spike created in a small initial volume by the incident particle. This minimum energy (E_m) required for bubble formation will be given in the next section.

2.4 Minimum Energy for Bubble Formation

The minimum energy (E_m) required for the formation of a critical nucleus is composed of a number of terms.

Norman and Spiegler (11) give E_m as:

$$E_m = \frac{4}{3}\pi R_c^3 \rho_v \Delta H + 4\pi R_c^2 \left(\sigma_l - T_l \frac{d\sigma}{dT}\right) + 2\pi \rho_l R_c^3 \dot{R}^2 + F \qquad (2.3)$$

where: R_c = critical bubble radius

ρ = density

ΔH = enthalpy change per unit mass

σ = surface tension

T = temperature

$\dot{R}$ = velocity of the bubble wall

F = viscous losses

and the subscripts v and l refer to the vapor and liquid states.

The first term on the RHS is the energy required to heat and vaporize the mass of liquid involved and expand it against the external pressure. The second term is the energy required to form a surface in the liquid. The third term is the kinetic energy given off to the liquid by the motion of the vapor wall. An expression for $\dot{R}$ is given (11) below

$$\dot{R} = \frac{4D}{R_c \left(\frac{\rho_v}{\rho_e}\right)^{\frac{1}{3}}} \tag{2.4}$$

where D is the thermal diffusivity.

The viscous loss term, F , has been shown ((11), (2), (12)) to be negligible compared to the other terms.

Deitrich (12) also uses equation (2.3) as the E_m required for bubble formation.

Becker (2) uses the following equation for E_m :

$$E_m = \frac{4}{3}\pi R_c^3 \rho_v \Delta H - \frac{8}{3}\pi R_c^2 \sigma + 4\pi R_c^2 \sigma - 4\pi R_c^2 \left(T\frac{d\sigma}{dT}\right) \tag{2.5}$$

Seitz (8), El-Nagdy (4), and Bell (5) use the E_m equation given below;

$$E_m = \frac{4}{3}\pi\left(R_c^2 \sigma + \rho_v \Delta H R_c^3\right) \tag{2.6}$$

Bell ((5) pg. 257) has also derived equation (2.6) from a purely thermodynamic viewpoint, which will be considered below.

Two systems are shown below. System I is liquid only, while system II contains a vapor bubble in the liquid. (See FIG.2.1 below) The energy required to go from system I to system II will be found.

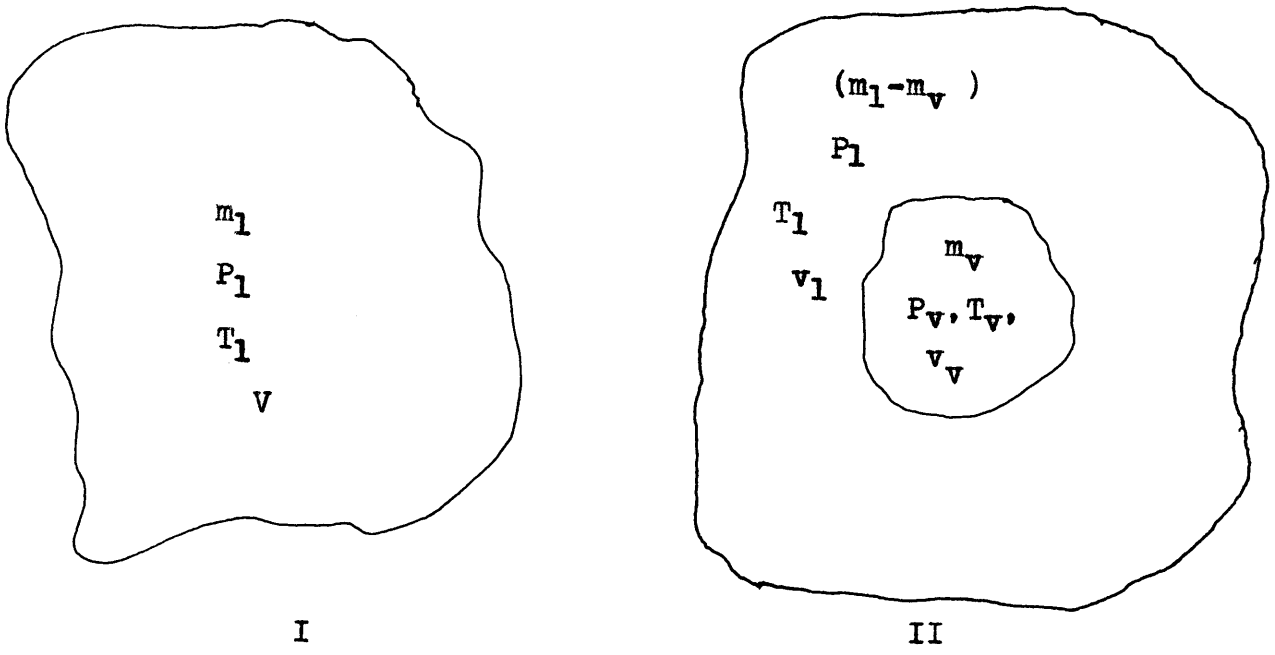


FIG. 2.1

The system parameters are defined as follows:

m = mass of the fluid

P = pressure

T = temperature

V = volume , v = Specific volume

The subscripts v and l again refer to the vapor and liquid states.

From the first law of thermodynamics

$$U_{II} - U_I = Q_{II} - W_{II} \qquad (2.7)$$

where U_x = internal energy of system x

Q_x = energy added to system x

W_x = work done by system x

The total energy terms are now converted to specific terms,

$$(m_\ell - m_v)u_\ell + m_v u_v - m_\ell u_\ell = Q_{II} - W_{II} \qquad (2.8)$$

The work done by the system (W_{II}) is composed of two parts: the work done to create the void, and the work done to form the surface. W_{II} is given by

$$W_{II} = P_\ell m_\ell (v_v - v_\ell) + A\sigma \qquad (2.9)$$

where σ = surface tension of embryo

A = surface area of the vapor embryo

Substituting equation (2.9) into (2.8) and rearranging terms,

$$u_v - (u_\ell + P_\ell v_\ell) = \frac{Q_{II}}{m_v} - P_\ell v_v - \frac{A\sigma}{m_v} \qquad (2.10)$$

Now $P_v v_v$ is added to both sides of equation (2.10):

$$(u_v + P_v v_v) - (u_l + P_l v_l) = \frac{Q_{II}}{m_v} - P_l v_v - \frac{A\sigma}{m_v} + P_v v_v \qquad (2.11)$$

From the definition of enthalpy:

$$h = u + Pv \qquad (2.12)$$

Substituting this into equation (2.11),

$$m_v (h_v - h_l) = Q_{II} + m_v v_v (P_v - P_l) - A\sigma \qquad (2.13)$$

Now, assuming (5) that the change in enthalpy (h_v-h_l) is approximately equal to the heat of vaporization:

$$h_{fg} \cong h_v - h_l \qquad (2.14)$$

Also, a number of substitutions can be made:

$$m_v = \frac{4}{3}\pi r^3 \rho_v \qquad (2.15)$$

$$A = 4\pi r^2 \qquad (2.16)$$

$$v_v = \frac{1}{\rho_v} \qquad (2.17)$$

where ρ = density

r = bubble radius.

For a critical embryo the equilibrium condition gives:

$$P_v - P_l = \frac{2\sigma}{R_c} \qquad (2.18)$$

Utilizing the above substitutions, equation (2.11) is reduced to:

$$Q_{II} = \frac{4}{3}\pi R_c^3 \left[\rho_v h_{fg} + \frac{\sigma}{R_c} \right] \qquad (2.19)$$

where Q_{II} is the energy which must be added to the system in order to create the critical embryo.(In other words, $Q_{II} = E_m$)

Equation (2.19) is exactly the same equation as the E_m equation employed by Seitz, El-Nagdy, and later by Bell. (Equation (2.6)) This is the equation which was used in the present work.

It was noticed that some investigators had included an additional surface tension term in their E_m equation (11), (3), (2). This term was of the form $T\frac{d\sigma}{dT}$

Seitz(8), El-Nagdy(4), and Bell(5) did not include this term. It was thus decided to see whether or not this additional term was significant. If this term were significant, then the original E_m equation

$$E_m = \frac{4}{3}\pi R_c^3 \left(\frac{\sigma}{R_c} + \rho_v h_{fg} \right) \qquad (2.20)$$

would be changed to

$$E_m = \frac{4}{3}\pi R_c^3 \left(\frac{\sigma - T\frac{d\sigma}{dT}}{R_c} + \rho_v h_{fg} \right) \qquad (2.21)$$

Calculations were made for each of the liquids studied to see whether or not this additional term would greatly affect the E_m equation.

The results of these calculations (TABLE 2.1) show that the $T\frac{d\sigma}{dT}$ term is negligible for the liquids studied (water, propylene glycol, nitromethane, benzene, acetone). The σ term in itself is only a small fraction of the E_m equation. The addition of the $T\frac{d\sigma}{dT}$ term does not significantly change this fraction.

This is true for the liquids studied in this work. But it does not necessarily hold for all liquids. In the case of liquid hydrogen and deuterium (used to a great extent in bubble chambers), this additional term is seen to significantly increase the contribution of the surface tension terms.

Therefore, before this additional term can be neglected, it must first be determined to what degree this term affects the E_m equation.

For the liquids studied in this work, this term was neglected, since it was proven to be negligible.

AFFECT OF ADDITIONAL SURFACE TENSION TERM

	A $\rho_v h_{fg}$ $\frac{lbf}{ft^2}$	B $\frac{\sigma}{r}$ $\frac{lbf}{ft^2}$	C % of Total B, % of A	D $\frac{\sigma - T\frac{d\sigma}{dT}}{r}$ $\frac{lbf}{ft^2}$	E % of Total D, % of A
WATER	180.1×10^3	2.56×10^3	1.4%	5.12×10^3	2.0%
PROPYLENE GLYCOL	98.05×10^3	0.365×10^3	0.3%	1.92×10^3	1.9%
NITRO-METHANE	193×10^3	0.88×10^3	0.4%	3.35×10^3	1.7%
ACETONE	93.2×10^3	0.34×10^3	0.36%	0.88×10^3	0.95%
BENZENE	122.6×10^3	0.73×10^3	0.59%	1.89×10^3	1.5%
HYDROGEN	41.97×10^3	2.83×10^3	6.74%	13.05×10^3	31.09%
DEUTERIUM	66.29×10^3	4.04×10^3	6.09%	19.90×10^3	30.02%

TABLE 2.1

2.5 Energy Balance Method of Bell

In his work Bell(5) postulated that the energetic particles involved in the nucleation process can be thought of as producing long cylindrical regions of energetic molecules along their path. Macroscopically, this cylindrical region can be thought of as a vapor cylinder. This vapor cylinder will have a diameter of approximately 100 Å ((5) pg63). A vapor cylinder in a liquid will tend to form into the more stable configuration, that of a sphere (or vapor bubble in this case). Since the total vapor track length is several orders of magnitude greater than the critical embryo diameter, it is highly unlikely that the total length of the vapor cylinder will reconfigure itself into a single vapor sphere. It is more likely that this long vapor cylinder will break up into smaller fragments, and that these fragments will reconfigure into individual embryos.

This process, as described by Bell(5), is shown schematically in FIG.2.2. Also shown is a typical energy deposition curve for a heavy charged particle interacting with matter. In the first part of the track the energy deposition to the electronic system of the stopping medium is the dominant energy transfer mechanism, while in the later part of the track the energy deposition to the medium through nuclear elastic

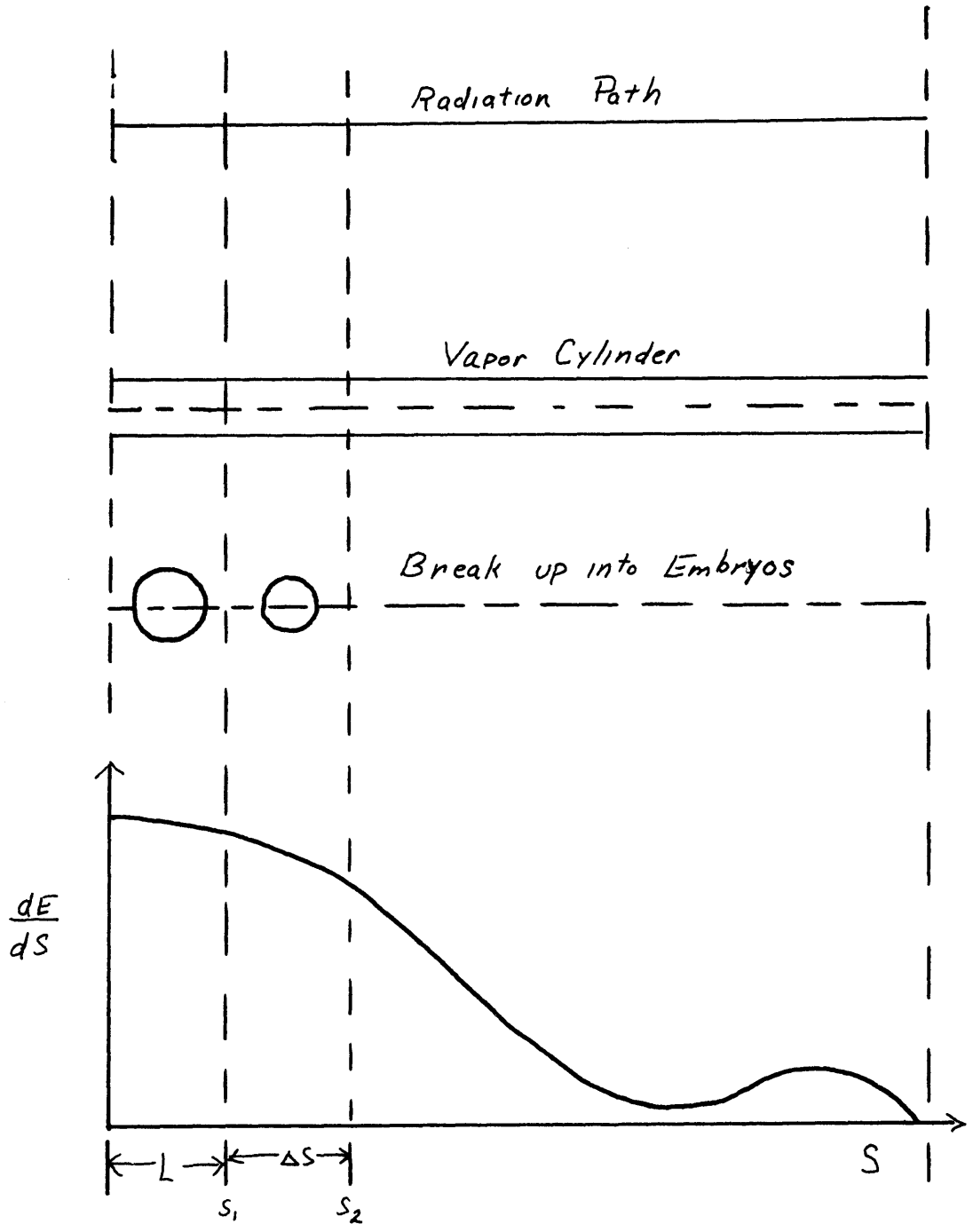

Radiation Path
Vapor Cylinder
Break up into Embryos
dE/dS
L
ΔS
S
S1
S2

FIG 2.2

collisions will be dominant.

If a section of the radiation particle track length is taken (say s_1 to s_2, or Δs) and assigned to one particular embryo, then the energy available to this embryo from the energetic radiation particle is the integral of the energy deposition rate, dE/ds , over the distance, Δs . This energy available to the embryo (or the energy lost by the particle) is given by

$$\Delta E = E(s_1) - E(s_2) = \int_{s_1}^{s_2} \left[\frac{dE}{ds}(E)\right] ds \qquad (2.22)$$

where $\left[\frac{dE}{ds}(E)\right]ds$ is a function of the particle energy at position s . But the energy deposition rate, dE/ds, is a function of energy, and not position(13). Therefore the integral in equation (2.22) cannot be solved directly. An approximation has been made ((5),74) for the average energy deposition rate:

$$\left(\frac{dE}{ds}\right)_{AVG} = \frac{1}{E(s_2) - E(s_1)} \int_{E(s_1)}^{E(s_2)} \left[\frac{dE}{ds}(E)\right] dE = \frac{E(s_1) - E(s_2)}{\Delta s}$$

OR

$$\frac{-\left[E(s_1) - E(s_2)\right]^2}{\Delta s} = \int_{E(s_1)}^{E(s_2)} \frac{dE}{ds}(E)\, dE \qquad (2.23)$$

The minimum superheat, or the threshold condition, for radiation-induced nucleation will be determined by the largest value of ΔE along the particle track. Assuming that the energy lost by the particle is equal to the energy made available to the embryo, and that the track breaks up in a uniform manner ($\Delta s = L =$ constant , (5) pg.74), then the maximum value for ΔE will occur where dE/ds is maximum, which is at the beginning of the radiation track in the stopping medium. (From $s = 0$, to $s = L$, as shown in FIG.2.2). $E(0)$ would simply be the initial energy of the energetic particle. If a value for $\Delta s = L$ is assumed, then equation (2.23) would contain only one unknown, $E(L)$.

Bell (5) assumed that this break up length is given by

$$L = ar^* \qquad (2.24)$$

where r^* is the critical radius of the embryo

"a" is a dimensionless parameter

At this point it will only be mentioned that Bell ((5) 203) postulated: $a = 6.07$. More will be said about this dimensionless parameter in chapter 6.

The basic concept behind the energy balance method is that the energy available from the radiation particle is equal to the energy required for bubble nucleation. The energy available, E_a, is given by

equation (2.23). The energy required for bubble formation was given by equation (2.6):

$$E_m = \frac{4}{3}\pi\left(r^{*2}\sigma + \rho_v \Delta H r^{*3}\right) \qquad (2.6)$$

There is another factor which must be added to the above equations. This factor is commonly referred to as an "energy sink", and arises from the formation of gas due to molecular dissociation in the stopping medium. In the liquids studied in this work, hydrogen is the gas produced by the dissociation process. The total energy loss for all the hydrogen formed in a track length L (or ar^*) is given by

$$\Delta E = \frac{Q \Delta E G(H_2) 10^6}{N_{AV}} \qquad (2.25)$$

where Q and $G(H_2)$ are constants, and ΔE is the same ΔE as in equation (2.23). This additional term is usually subtracted from the available energy, E_a , term.

The final form of the energy balance equation is thus:

$$\frac{4}{3}\pi r^{*3}\left[\frac{\sigma}{r^*} + \rho_v h_{fg}\right] = \Delta E\left[1 - \frac{Q G(H_2) 10^6}{N_{AV}}\right] \qquad (2.26)$$

where the LHS is the minimum energy required for bubble formation (E_m) and the RHS is the total energy available, E_a.

When equation (2.26) is satisfied, then the conditions for radiation-induced nucleation have been met.

Besides being an energy sink, the production of the hydrogen gas also affects the dynamic equilibrium of a critical embryo. The original mechanical equilibrium condition for a critical embryo is given by:

$$P_v - P_l = \frac{2\sigma}{r^*} \qquad (2.27)$$

Taking the hydrogen gas into account, equation (2.27) becomes:

$$P_v + P_G - P_l = \frac{2\sigma}{r^*} \qquad (2.28)$$

The effect of this hydrogen gas is to reduce the vapor pressure requirement inside the critical nucleus for a nucleus of a particular size, and thus reduce the superheat required for that critical nucleus. An equation for P_g (the gas pressure term) is ((5), pg122)

$$P_G = \frac{3 k T_v \Delta E G(H_2) 10^6}{4\pi r^{*3}} \qquad (2.29)$$

P_g cannot be calculated directly from equation (2.29), since r^* is a function of P_g (in addition to being a function of P_l), and ΔE is also a function of P_g, since $E(s_2)$ depends on r^* .

Before equation (2.29) can be solved, an expression must be found for dE/ds in equation (2.23). A number of energy deposition formulas have been suggested. The one which is most widely used is that given by Segre(13). This has also been modified for fission fragments. The equation is given below:

$$\frac{1}{N}\frac{dE}{ds} = \frac{4\pi e^4}{m_0 v^2}(Z_1)^2_{eff}\sum_i \nu_i Z_i \ln\left(\frac{1.123\, m_0 v^3}{(I_i/\hbar)e^2 (Z_1)_{eff}}\right)$$

$$+ \frac{4\pi e^4 (Z_1)^2}{v^2}\sum \nu_i \frac{Z_i}{M_i}\left[\ln\left(\frac{M_1 M_i v^2 (a_{12}^{SCR})_i}{(M_1+M_2) Z_1 Z_i e^2}\right)\right] \qquad (2.30)$$

The symbols used in equation (2.30) are defined below:

e = charge on an electron

N = number of molecules of stopping medium per unit volume (molecular density)

m_0 = electron mass

v = velocity of the particle going through the stopping medium

Z_1 = atomic number of the particle

Z_i = atomic number of the i^{th} atom in the stopping medium

ν_i = number of i^{th} atoms per molecule

I_i = mean ionization potential of the i^{th} component

M_1 = mass of the particle (amu)

M_i = mass of the i^{th} atom (amu)

$\hbar$ = Planck's constant divided by 2π

$(Z_1)_{eff}$ = effective charge on the particle.

An approximate relationship for the above term is given by Segre (13):

$$(Z_1)_{EFF} = (Z_1)^{1/3} \frac{\hbar V}{e^2} \qquad (2.31)$$

a_{1i}^{SCR} = impact parameter beyond which energy loss by the particle is effectively zero due to screening of nuclei by atomic electrons for the i^{th} atom.

An expression for the impact parameter is given by Claxton (16) :

$$a_{1i}^{SCR} = \frac{a_H}{\left[(Z_1)^{2/3} + (Z_i)^{2/3}\right]^{1/2}} \qquad (2.32)$$

where a_H is the radius of the first Bohr orbit for the hydrogen atom.

Equation (2.30) must be evaluated for each particular charged particle-stopping medium case. The solution to equation (2.30) will have the general form of

$$\frac{dE}{ds} = \rho_\ell \left[A \ln BE + \frac{C}{E} \ln DE\right] \qquad (2.33)$$

where A,B.C,D are constants

ρ_ℓ - is the density of the stopping medium

E is the energy of the radiation particle.

The first term in equation (2.33) represents the energy deposition to the electronic system of the stopping medium through ionization and excitation of the atoms, and the second term represents the energy deposition to the stopping medium through nuclear elastic collisions between the energetic particles and the nuclei of the stopping medium. In the cases considered in this work, the second term is negligible, and may be neglected. (This approximation is shown to be valid by Bell (5) pg. 117, and by Segre (13) pg 166.)

Equqtion (2.33) can thus be reduced to

$$\frac{dE}{ds} = \rho_\ell A \ln(BE) \qquad (2.34)$$

Equation (2.34) shows that the energy deposition rate, dE/ds , is a function of the particle energy, E. This fact was mentioned earlier in connection with equation (2.22).

Equation (2.34) is now substituted into equation (2.23), and the result takes the general form:

$$\frac{[E(s_1)-E(s_2)]^2}{E(s_1)\ln E(s_1)-E(s_2)\ln E(s_2)-B[E(s_1)-E(s_2)]} = Ca\rho_\ell r^* \qquad (2.35)$$

where B and C are constants

a = Bell's parameter

ρ_ℓ = density of the stopping medium

r* = critical radius of embryo

A substitution is now made for r*, using equation (2.28):

$$r^* = \frac{2\sigma}{P_v + P_g - P_l} \qquad (2.36)$$

It is now possible to calculate, using an iterative procedure, P_g.

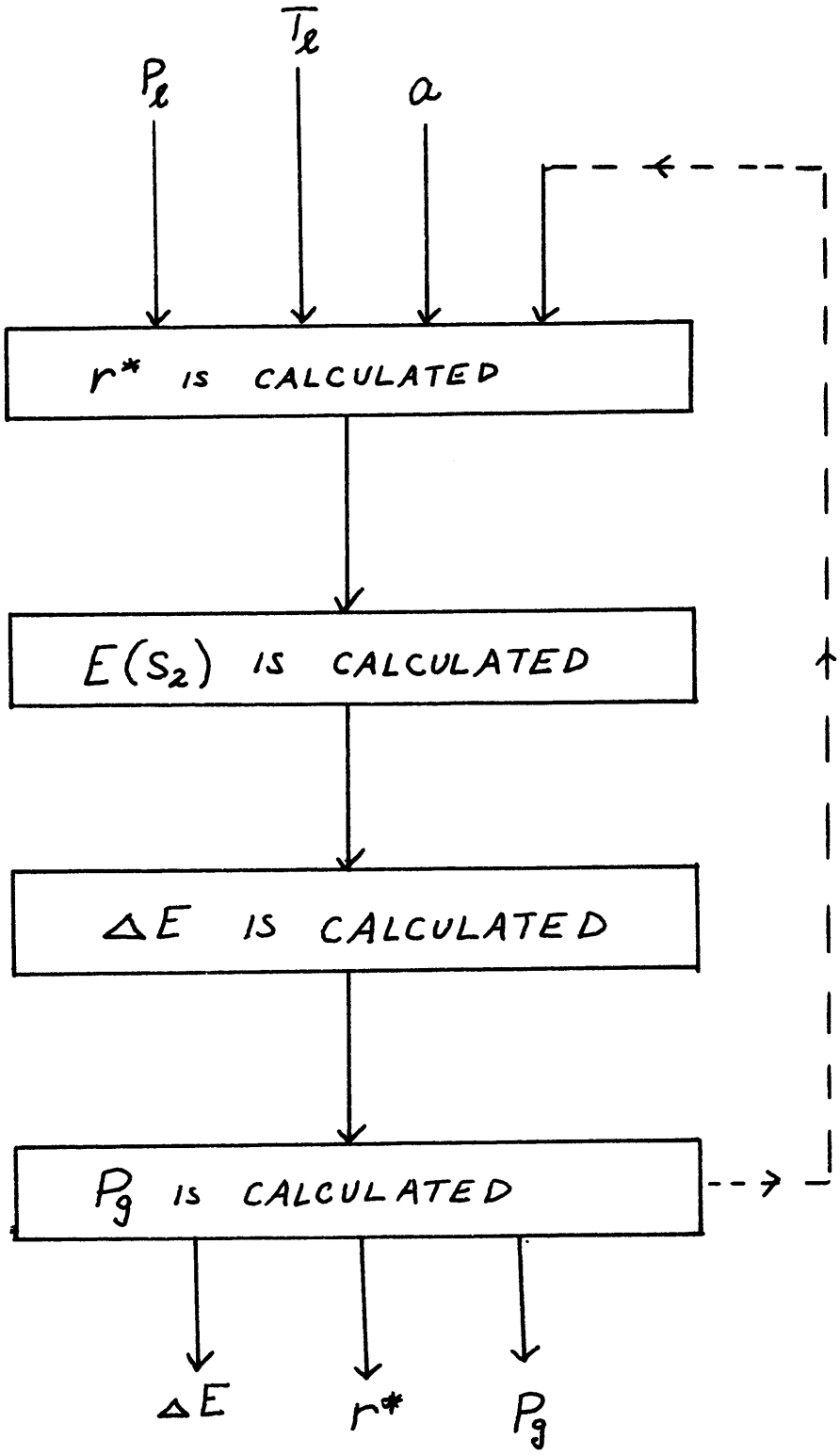
The first step in this procedure is to guess a value of P_g. r* is then calculated from equation (2.28) (P_v, P_l, and σ are known). The next step is to calculate $E(s_2)$ from equation (2.35). ΔE can now be found by recalling that

$$\Delta E = E(s_1) - E(s_2)$$

$E(s_1)$, the initial particle energy, is always known. P_g is now calculated using equation (2.29). If this calculated value of P_g is not equal to the original P_g guess, then another value of P_g is assumed, and the procedure is continued until the two values converge. FIG. 2.3 gives a simplified flow chart of this calculational procedure.

The convergence of this series will give correct values of P_g , r* , and ΔE , for a given set of values of P_l , a , and T_l .

Once P_g has been obtained, the superheat temperature can be obtained. A complete program has been developed by Bell((5), pg 122), which calculates the superheat

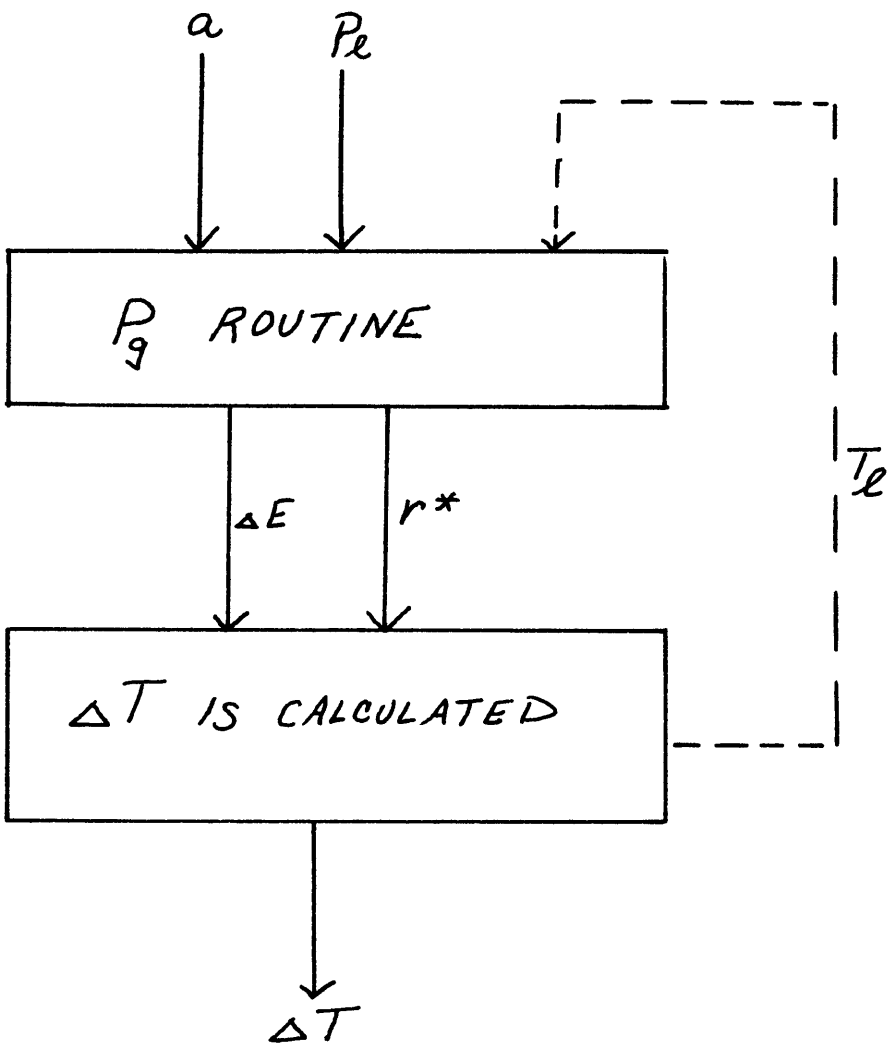


P_g ROUTINE

FIG. 2.3

threshold for various pressures and "a" values. It is an iterative procedure involving the system temperature, T_l . A brief description of the program operation will be given.

There are three independent variables: the system pressure, P_l, the system temperature, T_l , and the parameter "a" . P_l and "a" are considered independent parameters. The first step of this procedure is to select an initial value of T_l, for a particular set of P_l and "a" values. This temperature value is then fed into the P_g procedure mentioned earlier, (the P_g procedure can be considered a subprogram of the entire calculational method for obtaining the threshold superheat.) The convergence of the P_g routine gives the corresponding values of P_g, r^* , and ΔE , for the set of chosen variables P_l , "a" , and T_l . Using these values of ΔE and r^* , the LHS and RHS of equation (2.26), are calculated. If equation (2.26) is not satisfied, a new value of T_l is chosen and the process is repeated until the energy balance criteria is met. When this criteria is met, the T_l obtained is the superheat temperature corresponding to the threshold condition. FIG. 2.4 gives a flowchart of the entire computational process for obtaining the correct T_l value.



THRESHOLD SUPERHEAT CALCULATION

FIG. 2.4

2.6 Fission Fragments and Fast Neutrons

In the case of a fission fragment, the fragment itself can directly produce the highly energetic region in the medium which is necessary for radiation-induced nucleation. This is not true of a neutron. The neutron interacts with the nucleus of an atom, through elastic scattering, and transfers some of its kinetic energy to the nucleus, thus producing a charged primary knock-on atom (PKOA). This PKOA then deposits its energy in a manner similar to the fission fragment.

The only change which this introduces into the computer program, is in the initial energy determination. In the case of fission fragments it is known (14) that the most probable fission fragment pair which results from this fission process has the following characteristics:

Light fragment	Heavy fragment
A = 97	A = 138
$E(s_1)$ = 95Mev	$E(s_1)$ = 67 Mev
Z = 38	Z = 54

If the energy deposition rates of these two fragments are compared over the track length from s=0 to s=ar* (this is the portion where the largest dE/ds will be obtained) then it is seen that the lighter fragment has the higher dE/ds, and will give a higher ΔE.

Therefore, for the threshold condition, only the light fragment need be considered. Thus, for fission fragments the initial energy is known.

The initial energy of the PKOA must be calculated. Since the energy of the neutron is known, the energy of the PKOA can be calculated from elastic scattering theory (15):

$$E(0)_{PKOA} = \tfrac{1}{2}E_n\left[1-\left(\frac{A-1}{A+1}\right)^2\right](1-\cos\theta) \qquad (2.37)$$

where E_n is the neutron energy

A = atomic weight of the PKOA

θ = the angle the neutron is scattered in the center of mass frame of reference.

For a maximum E(0), $\cos\theta$ is taken as zero.

This is the only important difference in the computational procedure for the threshold values.

Chapter 3

Experimental Apparatus and Procedures

3.1 Apparatus Modifications

The experimental apparatus was the same as that used by Bell (5) and Tso (6), with a few modifications.

The original test chamber is shown in Fig. 3.1. The test bubble, X , is suspended in the suspension fluid,H, (Dow Corning 550 fluid for the case of water and propylene glycol) and covered with the cover fluid I (Nujol, a heavy mineral oil, for the case of water and propylene glycol). The test chamber, A , is a cylindrical (10"x 3" diameter) steel chamber. The test bubble is visually observed through the window B . The bubble temperature is recorded by two Chromel-Alumel thermocouples (K and L). The chamber is heated by two electrical heaters (Mand N). The cover fluid is introduced into the test chamber from reservoir E through the cover fluid outlet tube J . The suspension fluid is introduced from the bottom of the test chamber from the reservoir Q. There are two cooling devices in the test chamber. One is the condenser, F , which condenses any water vapor in the air space left by previous boiling. The condensate is collected in a container and discharged through a tube into reservoir R . The other cooling device is the convection generator G . This serves not only as a cooling device

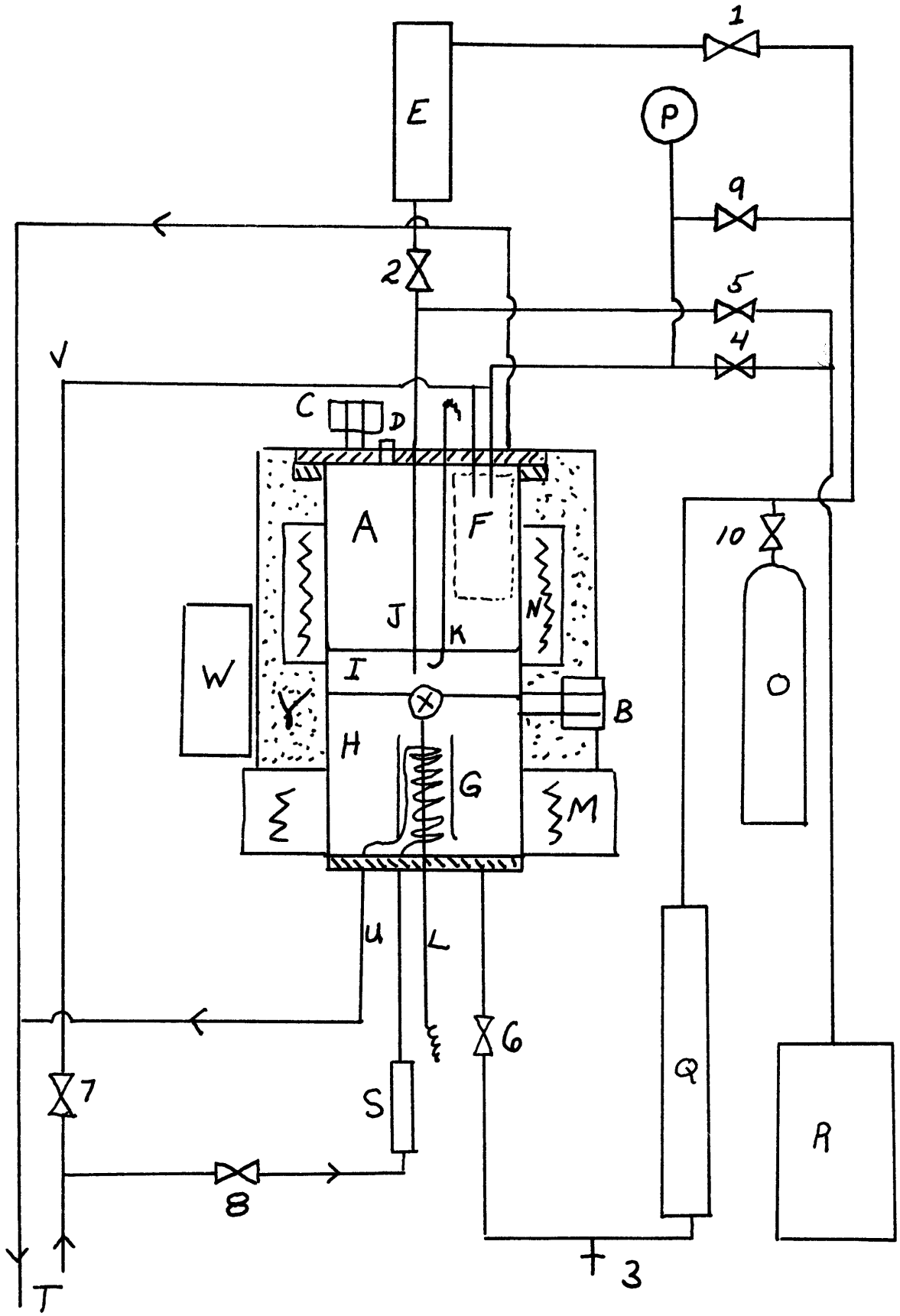
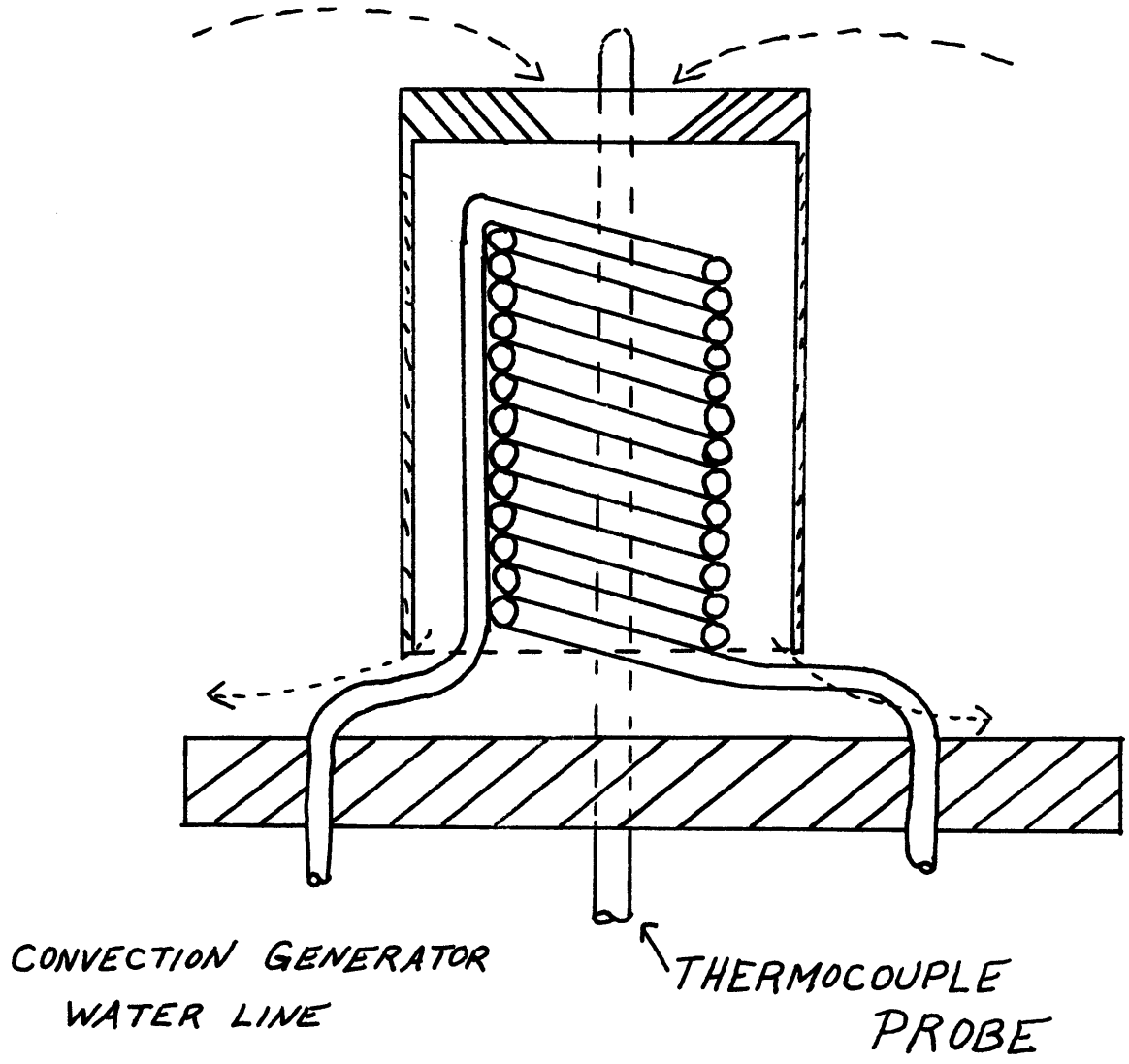


FIG. 3.1

Number 1 to 10	valves
A	boiling chamber
B	observation window
C	light window
D	bubble entrance
E	cover oil reservoir
F	condenser
G	convection generator
H	supporting oil
I	cover oil
J	cover oil outlet
K	top thermocouple
L	bottom thermocouple
M	bottom heater
N	top heater
O	air pressurizer
P	pressure gauge
Q	supporting oil reservoir
R	waste tank
S	flow indicator
T	supply water line
U	convection generator cooling lines
V	condenser cooling lines
W	neutron source
X	water bubble
Y	fibre-glass insulation

when the chamber is being cooled down, but it serves the important function of keeping the test bubble centered during the experimental run. Since this convection generator is such an important part of the apparatus, it is shown separately in FIG. 3.2 . The manner in which the bubble is kept centered is also shown in this figure. The cooling water through the convection generator coils causes a convection current which circulates the supporting fluid such that the flow is radially towards the top of the generator, down through the generator, and radially out at the bottom. In this way the bubble is kept radially in place in the test chamber. The vertical position of the bubble is set by adjusting the supporting oil level.

Two important modifications were made. The first, and most significant, was a change in the method of illuminating the test chamber. In the original design a high intensity lamp was placed above window C, and this was considered adequate. It provided sufficient light to see the test bubble, but it was of considerable strain to the eyes, thus affecting the ease and accuracy of obtaining the data, since all data was visually obtained. The modification which was made placed the light source directly into the chamber, giving a far better illumination of the test bubble. FIG. 3.3a shows the original method, and FIG.3.3b shows the final illumination method. (Only the section of the chamber which supports the lighting system is shown in FIG.3.3)



CONVECTION GENERATOR

FIG. 3.2

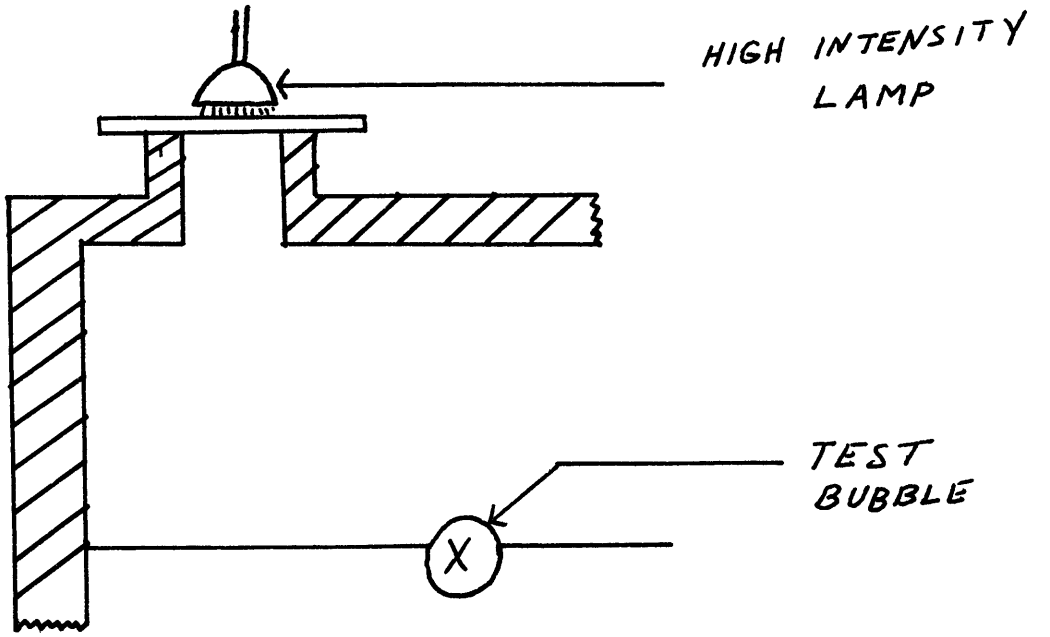


FIG. 3.3a

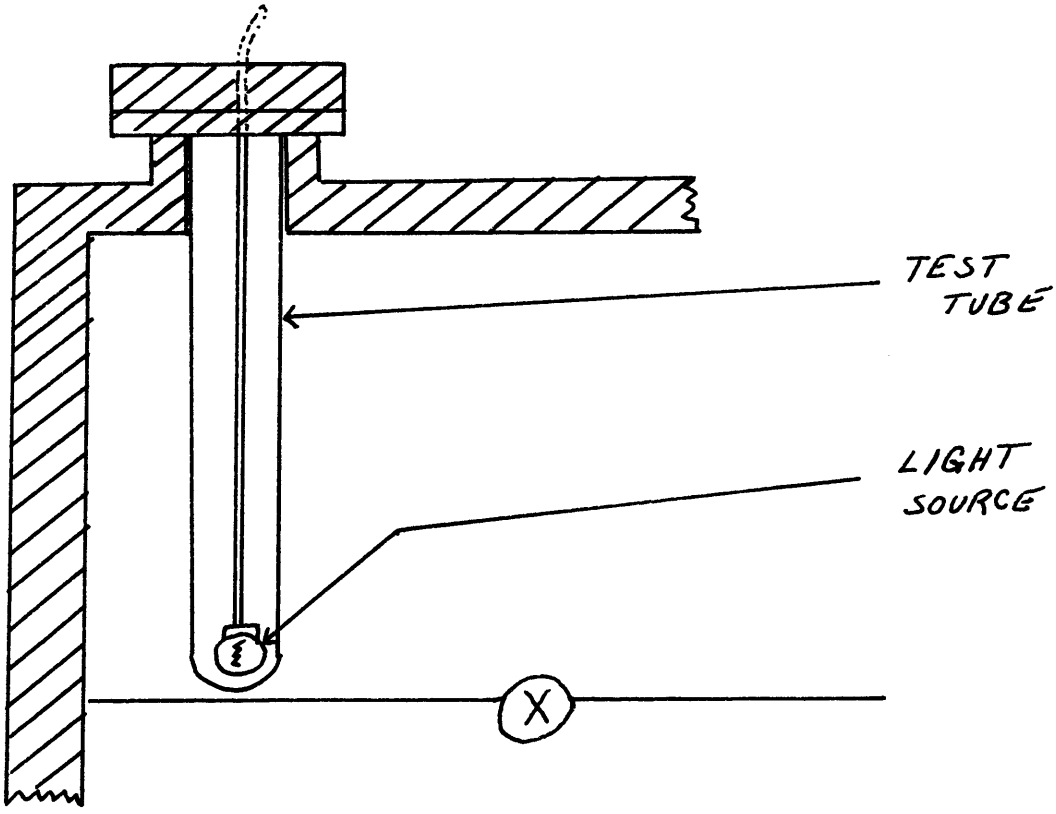


FIG. 3.3b

This modification required a great amount of work. Since it was not desired to construct a new chamber, it was necessary to work through the existing window C. This created a number of problems, the greatest being that of obtaining a pressure seal at window C. The problem was eventually solved by using the combination of metal rings and hard rubber gaskets to sandwich in the test tube flange. (see FIG.3.3b) Pressures of up to 100 psig were obtainable with this design.

The other modification was to introduce a mirror system for the observation of the bubble. The original design required the experimenter to place his eye directly in front of observation window B. This was an extremely uncomfortable position, and had to be maintained for the length of the experimental run which often exceeded a half-hour.

The mirror system (shown in FIG.3.4) permitted the experimenter to observe the bubble from a distance, and made it easier to monitor all gauges and recorders indicating the temperature and pressure of the test chamber.

The other equipment which should be mentioned is the radiation source, a 5 curie Pu-Be source, and its associated shielding.

The source itself is a small cylindrical element, which is contained in a rectangular aluminum container. This is attached to a masonite block. This entire unit is kept in a shielded storage container until the source

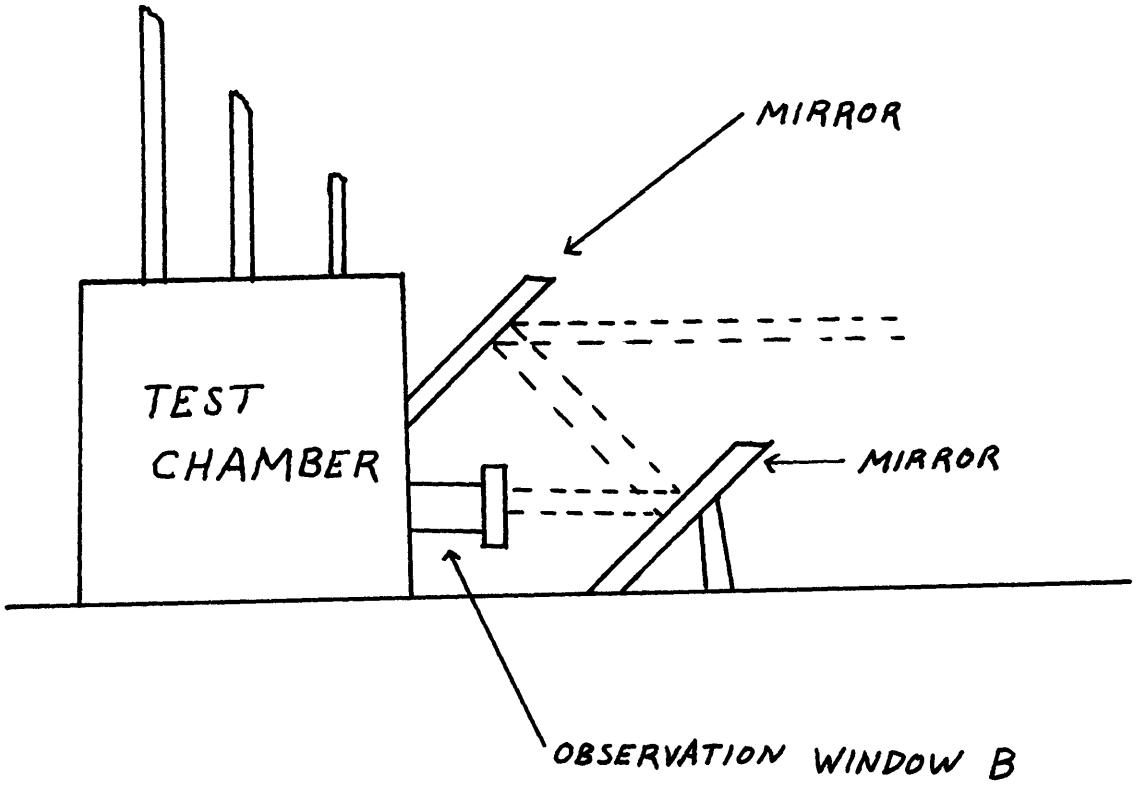


FIG. 3.4

is needed. It is then placed in a shielded housing which has been constructed adjacent to the test chamber. (The source and housing are shown in FIG. 3.5)

The pressure gauge P was tested for accuracy with a dead weight tester, and it was found that the gauge read approximately 3 psi too low. The calibration curve is shown in FIG.3.6 .

3.2 Experimental Procedure

The procedure for a typical experimental run will be outlined to describe the operational procedure.

Before beginning any experiments with a new test liquid, it is important to thoroughly clean the inside of the test chamber. (Washing with water and then with acetone is recommended.).

The operational procedure is outlined below:

1) Introduce the suspension fluid by opening valves 6 and 10, and closing all others. This will force the suspension fluid from the reservoir Q , into the test chamber. The supporting fluid level should be brought to near the top of window B (as shown in FIG.3.1). Once the desired level is reached, valves 6 and 10 are closed.
2) The cover fluid is next let into the chamber by opening valves 1,2, and 10. Approximately

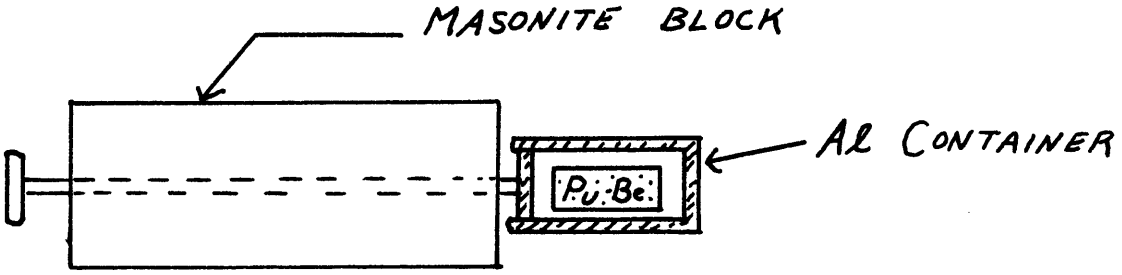


FIG. 3.5.a

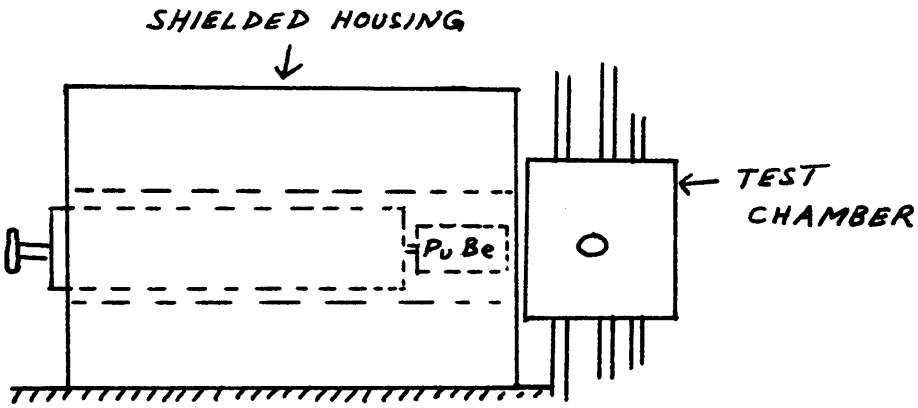


FIG. 3.5 b

CALIBRATION CURVE FOR PRESSURE GAUGE

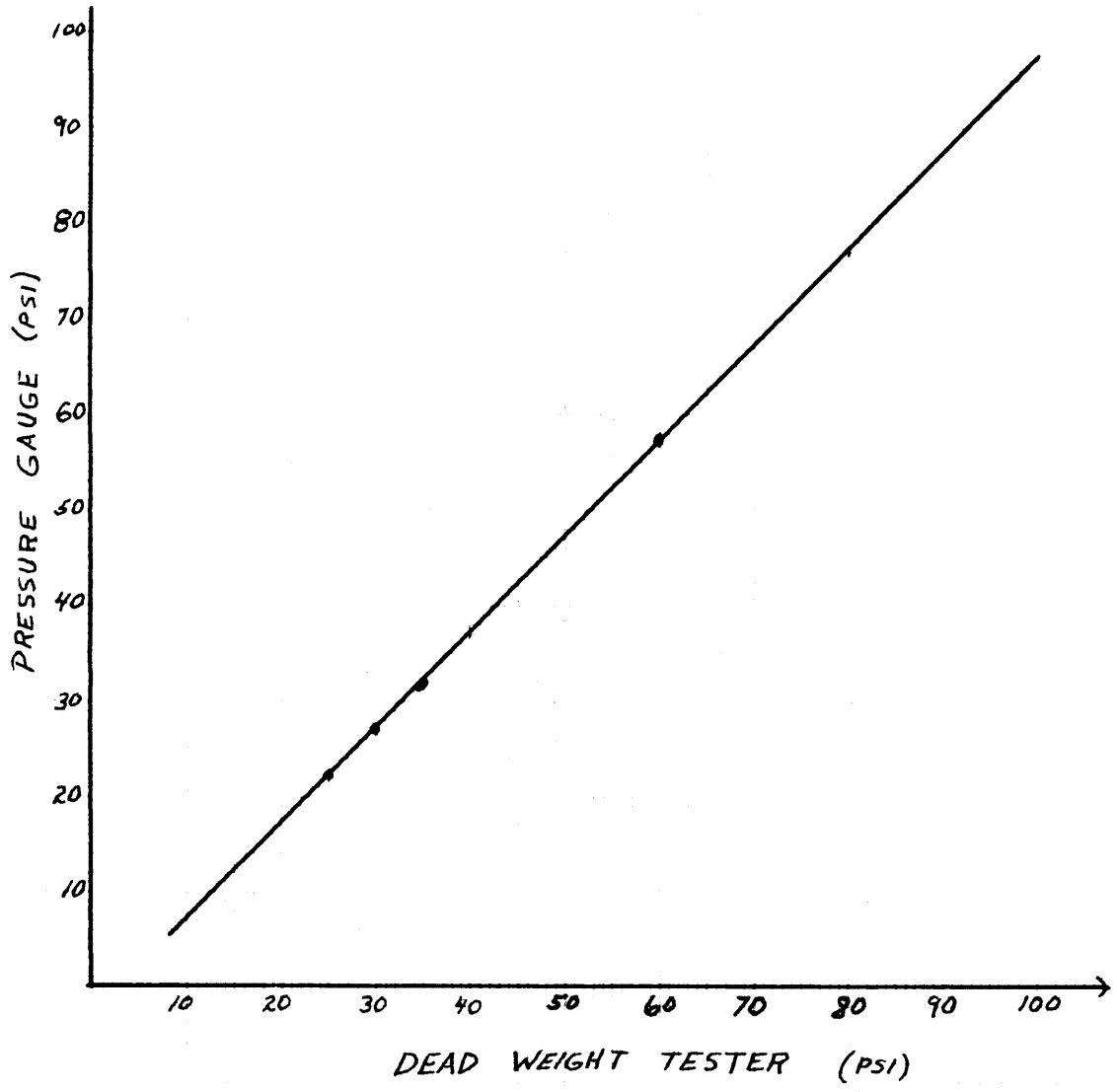


FIG 3.6

an inch of cover fluid is sufficient. When the desired level is reached, valves 1,2, and 10 are closed.

3) Valve 8 is opened to allow the convection generator to start flowing.

4) The test bubble is then introduced through opening D , by means of an eye dropper. The test bubble is placed above the center of the convection generator.

5) After the bubble has been introduced, opening D is closed, and all valves, except 8, are closed.

6) Valves 9 and 10 are then opened, with valve 10 being adjusted to the desired pressure.

7) Heaters M and N are turned on, and the temperature is constantly monitored through thermocouples K and L .

8) Once nucleation is observed, or the run is to be terminated, both heaters, M and N , are turned off, valve 7 is opened, and valve 8 is turned on full (if it were not already fully opened). The chamber is then left to cool down. The time required for it to cool down depends on the final temperature which was reached during the heating process. A final temperature of 500 F will require approximately 45 minutes.

9) When the chamber has cooled down the top layer of liquid must be drawn off, since the test bubble will have broken up and must, in any case, be removed and a new test bubble put into the chamber. This is done by raising the liquid level (by opening valves 6 and 10) until all bubbles are above the opening of the cover oil outlet tube J. All valves are then closed. Valves 9 and 10 are opened, thus pressurizing the chamber. Then valve 5 is opened. This will draw off the top layers of fluids and eject them into reservoir R .

When this has been completed all valves are closed, and the system is ready for another experimental run, beginning with step 1 of this procedure.

3.3 Experimental Difficulties

While running the experiment a number of problems may arise which can create serious difficulties, leading to a premature termination of the experiment.

One problem to be avoided is that of mixing of the cover and suspension fluid. This can occur if valve 6 is opened too quickly or too much, causing the suspension fluid to shoot up into the cover fluid. Also, if reservoir Q is empty and valve 6 is opened, air will shoot through the liquid and cause violent agitation and mixing of the

fluids. The physical properties of the cover and suspension fluids are such that once they are mixed they form a cloudy solution and are very difficult to separate. When mixing occurs the chamber must be emptied and cleaned, and new fluids introduced. It is therefore important that reservoir Q always be filled. The same is true for reservoir E .

Another point to remember is that when cooling down the chamber after a high pressure run, the pressure must not be suddenly reduced. This will cause immediate and violent boiling which will thoroughly mix the fluids. The chamber should thus be cooled down to a temperature below the boiling point of the liquid at atmospheric pressure, and then the pressure can be released.

Another problem might be a flow blockage problem with the convection generator. The tubing used in forming the convection generator is a very small diameter refrigeration tubing. Ordinary tap water is used as the coolant water, and this contains a small amount of chlorine which tends to corrode the inside of the tubing. There are a number of bends in the small diameter tubing which can become blocked due to small particles already existing in the water or produced by corrosion. If the tubing is not periodically cleaned (a weak solution of sodium hydroxide is recommended) the corrosion deposits will soon build up and block the flow. This was the case in the present work. The corrosion deposits were too

thick to be able to be cleaned, so a new convection generator had to be constructed.

During the propylene glycol experimental runs, a change in color of the test liquid was seen to occur at around 300 F. This change is believed to be a chemical characteristic of the liquid. (This characteristic is also seen in other organic liquids.). Also, small dark particles were noticed in the test chamber after a number of experimental runs had been made with propylene glycol. These are believed to be due to residual uranium nitrate particles which remained in the chamber after the propylene glycol test liquid boiled. (The uranium nitrate was introduced to the test liquid for the purpose of providing fission fragments. See section 3.5 for a further discussion on this matter.) These particles required additional cleaning of the chamber.

3.4 Indication of Nucleation

In all previous work on radiation-induced nucleation, the actual production of vapor bubbles in the test liquid proved the nucleation event. In the experimental procedure developed by Bell(5), nucleation is observed through the jumping or break-up of the test bubble. That is, when the bubble is seen to suddenly jump, nucleation has occurred.

For fission fragments at low pressures there is no difficulty at all in observing when nucleation occurs.

The nucleation event at these conditions causes the bubble to actually break up or "explode". At high pressures, nucleation due to fission fragments will cause the bubble to jump suddenly. This is easily seen since it is a fairly large jump. The first jump is the important one, since this gives the minimum superheat value. If the temperature is raised beyond this point, the jumps will be more frequent and become more violent.

For the case of fast neutrons it is more difficult to observe the nucleation event. To begin with, higher temperatures are required, which immediately lengthen the experimental run. At low pressures the nucleation event causes the test bubble to jump in a manner similar to the case of fission fragments in the test fluid. But at high pressures the test bubble jumps only very slightly, and extremely careful observation is required in order to notice this small movement of the bubble. This fact makes it difficult to obtain data for fast neutron-induced nucleation at high pressures.

3.5 Fission Fragments in a Test Liquid

Fission fragments are introduced into the test fluid dy dissolving a minute quantity of Uranium Nitrate ($UO_2(NO_3)_2 \cdot H_2O$), in the test liquid. The amount of Uranium nitrate dissolved can vary. For the case of water, the concentration used was : 0.0087 gm of uranium nitrate to 1.0 gm of water. For the case of propylene

glycol , 0.0029 gm of uranium nitrate to 1.0 gm of test fluid was used.

The fast neutrons from the 5 curie Pu-Be source will cause fission events in the uranium nitrate, thus creating the desired fission fragments.

Chapter 4

Fluid Selection

4.1 Background Information

As mentioned previously, the experimental technique employed by Bell consisted of suspending the test liquid in two other fluids. This gives the important advantage of having the test liquid free from any surface effects.

The test liquid used by Bell(5) and Tso(6) was water. The supporting fluid was a Dow-Corning 550 silicone fluid. The cover fluid was a heavy weight mineral oil, which has the trade name of Nujol. Both experimental and analytical work was done with water.

Tso(6) did some preliminary calculations for benzene as a test fluid, exposed to Pu-Be neutrons. A literature search found benzene(4), acetone(4), and diethyl ether (2) used as test liquids in radiation-induced nucleation studies. It was thus decided to confirm and extend the work of these other investigators, and to obtain analytical results using Bell's(5) constant "a" theory.

4.2 Test Liquid Properties

A number of physical properties are required for the computation of the superheat temperature. These properties are listed below:

1.) Vapor Pressure

2.) Liquid Density

3.) Vapor Density

4.) Surface Tension

5.) Heat of Vaporization

Data for each of the above physical properties must be known at a number of temperatures, since the calculational procedure requires an empirical relationship between the physical property and the temperature.

This empirical relationship is obtained by feeding the data for various temperatures into a polynomial regression computer program. This program will give an expression of the form:

$$P = A + BxT + CxT^2 + DxT^3 + \ldots\ldots \qquad (4.1)$$

where: P = the value of the physical property

T = temperature at which the property is to be evaluated

A,B,C,D,.... are constants determined by the program.

The polynomial regression program is given in Appendix C.

In addition to benzene, acetone, and diethyl ether, a search was also conducted to find the required properties for methyl alcohol, ethyl alcohol, nitromethane, and propylene glycol.

All required properties were eventually found, and the empirical relationships,along with graphs of the physival property versus the temperature, are given in Appendix A .

4.3 Compatibility Requirements

A simple schematic drawing of the configuration of the liquids is shown in FIG.4.1 below;

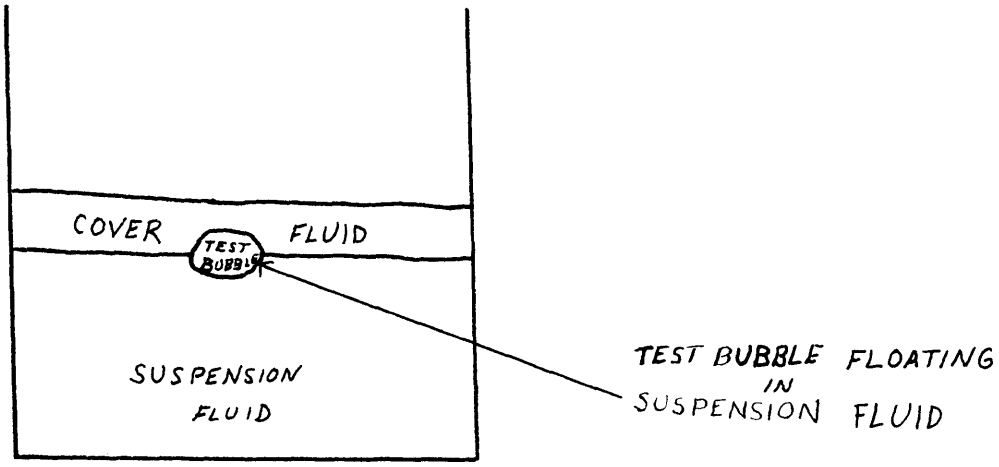


FIG 4.1

The three fluids must exhibit the following characteristics:

1.) The three fluids must be immiscible with each other. That is, they cannot be soluble with each other.

2.) The relative densities must be such that the supporting fluid has the highest density, the cover fluid the lowest,and

the test fluid density must lie between the two.

3.) The boiling points of the cover and supporting fluids must be greater than the boiling point of the test fluid. The lower the test fluid boiling point, the easier the experimental work.

4.) The surface tension difference between the test fluid and the other two fluids must be fairly large, with the test fluid having the largest surface tension value.

The fourth factor listed above is not that obvious at first, but it is one of the most important factors affecting the choice of the three liquids.

4.4 Difficulties in Fluid Selection

Benzene and Acetone were originally planned to be used as the test fluids, since some work had already been done on these fluids (4). But benzene and acetone were not able to be used with this three liquid configuration. The problems presented were twofold:

1.) The densities of benzene and acetone are very low:

- benzene density 0.87901 gm/cm^3 (20 °C)
- acetone density 0.78998 gm/cm^3 (20 °C)

2.) Benzene and acetone are two of the most universal solvents in use. In other words, they are soluble in almost any liquid.

These two factors seemed to work together. The few liquids which are insoluble with benzene or acetone all have higher densities. This eliminates any possibility for a cover fluid, which must have a lower density.

Evidence of this solubility problem can be seen in any solubility chart. TABLE 4.1 is a small section of a large solubility chart (reference (17) pgs. 58-60), and it is seen that there are indeed few liquids which are insoluble in benzene and in acetone. The liquids which were found to be immiscible with acetone and benzene are listed in Table 4.2 and 4.3 , along with their density. In all cases, those liquids which were immiscible with benzene or acetone had higher densities . An extensive literature search (and consultations with graduate students in the organic chemistry department at M.I.T.) failed to produce a suitable cover fluid for either benzene or acetone.

Therefore, only analytical work was able to be done with benzene and acetone. However the data of El-Nagdy (4) is later compared with these analytical results to determine the "a" value predicted by this data.

	ACETONE	BENZENE
Acetone		M
Isoamyl acetate	M	M
Benzene	M	
Chloroform	M	M
Diethylenetriamine	M	M
Ethyl alcohol	M	M
Ethyl Benzoate	M	M
1,3-Butylene glycol	M	I
Ethyl ether	M	M
Methyl isopropyl ketone	M	M
Triethylenetetramine	M	M
Glycerol	I	I
Propylene glycol	M	I
Trimethylene chlorohydrin	M	M
Carbon tetrachloride	M	M
Nitromethane	M	I
Diethyl ether	M	M
Pyridine	M	M
Diethyl cellosolve	M	M
Triethyl phosphate	M	M

M = miscible I = immiscible

TABLE 4.1

TABLE 4.2

Liquids immiscible with benzene

Liquid	density gm/cm^3 (20°C)
1,3-Butylene glycol	1.005
3-chloro-1,2-propanediol	1.326
ethylene glycol	1.1135
glycerol	1.261
1,2-propanediol	1.036
1,3-propanediol	1.053
diethanolamine	1.089
formamide	1.133
hydroxyethyl-ethylenediamine	0.980
nitromethane	1.1138

TABLE 4.3

Liquids immiscible with acetone

Liquid	density gm/cm^3 (20°C)
glycerol	1.261

A search was conducted to find a test liquid other than water, and supporting liquids which would be compatible with it. This is a difficult and time consuming procedure, since there are four independent variables which must be taken into account (miscibility, density, boiling point, and surface tension).

The easiest method of search is to begin with the miscibility condition. This is the quickest way to eliminate most of the unsuitable liquids. The first step would be to choose liquids which are immiscible with a large number of other liquids. (The miscibility charts given in references (17)-(21) are recommended for this purpose.)

After obtaining a number of candidates from the miscibility criteria, the boiling points should be checked. The test liquid should have as low a boiling point as possible. (Reference (22) gives the boiling points of most of the commonly used liquid solvents conveniently in order of increasing boiling point.) The possible choices are then checked as to densities. The test liquid must have an intermediate density. (Reference (22) also gives the densities of a large number of liquids in ascending order.)

In addition to the organic liquids there exist some light weight mineral oils which might be able to be used as cover and stopping fluids. These are for the most part clear liquids with high boiling points, and

are immiscible in many organic liquids. But even though they are referred to as light-weight oils, their densities are not much less than that of water. (Nujol has a density of 0.886 gm/cm^3 at 20°C, which makes it heavier than either benzene or acetone.)

Upon completion of the preceeding analysis, a number of possible choices were available. The exact effect of surface tension was not known at this time. Small quantities of the most likely candidates were obtained, and compatibility tests were made. These compatibility tests simply consisted of placing the supporting and cover fluids in a beaker (a similar arrangement as in the test chamber) and then introducing the test fluid. This is an exact simulation of the manner in which it is done in the test chamber.

Table 4.4 gives a partial list of the results observed from the compatibility tests described above. In many cases there resulted what was termed a "spreading effect", where the test liquid did not remain in the shape of a bubble, but spread out over the supporting fluid. This effect is shown in FIG. 4.2.

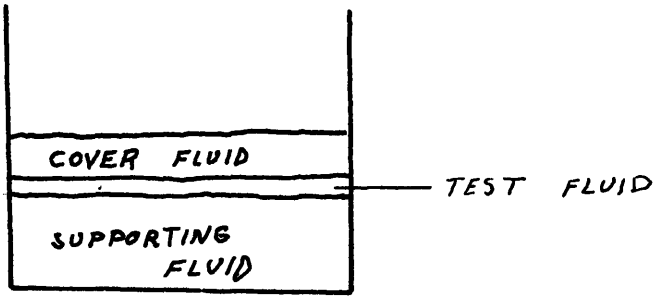


FIG. 4.2

	SUPPORTING FLUID	TEST FLUID	COVER FLUID	RESULT
	(surface tension in dynes/cm at 20°C)			
(1)	Glycerol 63.3	Nitro-methane 37.48	Nujol 23.0	Spreading of the test fluid.
(2)	Ethylene-glycol 46.49	nitro-ethane 32.66	Nujol 23.0	Spreading of the test fluid.
(3)	Glycerol	1,2-dichloro-ethane 32.23	nujol	Test bubble slightly soluble when heated.
(4)	Glycerol	propylene-glycol 36.9	Nujol	Bubble does not remain in bubble shape, spreads.
(5)	Glycerol	salicyl-aldehyde —	ethylene-glycol	test bubble soluble when heated
(6)	DC FS 1265 fluid 26.1	nitro-methane	Nujol	test bubble slightly soluble when heated.
(7)	DC 550 fluid 23.50	propylene-glycol	Nujol	Stable bubble

TABLE 4.4

The surface tension effect is apparent from these results. In the fourth test listed the supporting fluid, glycerol, has a high surface tension (63.3dynes/cm at 20°C), while the test fluid, propylene glycol, has a much lower surface tension (36.9 dynes/cm at 20°C). The result of this test was the spreading of the test fluid over the supporting fluid as shown in FIG.4.2 . In the seventh test listed, the high surface tension glycerol was replaced with a low surface tension (23.5 dynes/cm at 20°C) DC 550 silicone oil. The result of this test was the successful configuration shown in FIG. 4.1 . In both of the cases mentioned the surface tension difference was large, but for a successful configuration it is not only necessary that there be a large difference between surface tensions, but also that the test liquid have the highest surface tension.

In the experimental work with water as the test fluid, the water test bubble showed exceptional stability in the supporting (DC550 fluid) and cover (Nujol) fluids. The surface tension of water is found to be 72.0dynes/cm at 20°C . This tends to support the conclusion arrived at above.

Thus a compatible set of fluids was found:

cover fluid : Nujol

test fluid : propylene glycol

supporting fluid : Dow Corning 550 fluid.

Experimental work was then done using this test fluid. (Due to time limitations, the search for more compatible fluid sets was not continued to find another possible test fluid.)

4.5 Discussion of Surface Tension

Since the surface tension plays such an important role in the selection of a compatible fluid set, a few words on the nature of surface tension are in order.

Surface tension (σ) is usually defined from a molecular viewpoint. An individual molecule in a body of liquid is subject to forces due to its neighboring molecules. (Theory indicates that these are electrical forces (23)) These forces will fluctuate due to molecular agitation, but when averaged over a finite period, their effect will be zero, so that a molecule can move about in a liquid without doing any work against these forces (24). In other words, the resultant average forces are equal in all directions inside a body of liquid.But when a molecule comes to the surface of this body of liquid, the molecule must ultimately move against unbalanced forces, since it will no longer be surrounded symmetrically by other molecules. At the surface, therefore, there will be an uneven distribution of forces acting essentially normal to the free surface of the liquid. (This will be normally inward, since the electrical force is essentially an attractive force (25)). There is thus a pull toward the interior

of the body exerted on the surface layer.

Since this surface force is uniform due to the averaging effect of the large number of molecules per unit of surface area, it tends to produce a surface with the smallest possible area. (If the liquid is free from gravitational effects, a spherical shape would result.) This inward normal pull is usually defined as the surface free energy. And to extend this surface, work must be done equivalent to this energy. This is usually referred to as the surface tension.(This term is not totally correct, but the idea of a tension in this free surface of a liquid is familiar as an explanation of the tendency of a liquid surface to assume the form having a minimum area.)

The units of surface tension are: dynes/cm , lbf/ft , or ergs/cm^2 .

Relationships between the surface tension and other physical properties will now be considered.

The first important fact is that as the temperature of the liquid is increased, the surface tension decreases. This is due to the fact that the increase in temperature gives the molecules in the liquid more energy, and thus larger fluctuations, and, since the attractive forces are close-range forces, they will decrease with an increase in the distance between molecules. At the critical temperature the surface tension is zero.

A number of relationships are given between the temperature of a liquid and its surface tension. These are all of the general form

$$\sigma = A - BxT \qquad (4.2)$$

where:

σ = surface tension

T = temperature at which the surface tension is to be calculated

A and B are constants which vary with the liquid. There is also a relationship between the viscosity and the surface tension. An equation relating these two parameters has been formulated by Buehler (27), and is given below:

$$\sigma^{\frac{1}{4}} = \frac{\log(\log \eta) + 2.9}{I/P} \qquad (4.3)$$

where: σ = surface tension

η = viscosity in millipoise

I/P = a constant which varies for different liquids.

A relation between surface tension, density, and temperature has been given by Macleod(34) :

$$\frac{\sigma}{(D-d)^4} = \text{constant} \qquad (4.4)$$

where; σ = surface tension

D = liquid density

d = vapor density

Very recently there has been developed a correlation between surface tension and the dielectric constant, and surface tension and the index of refraction (28). The relation between the surface tension and the dielectric constant is shown in equation (4.5)

$$\sigma = 165.0\left[(\epsilon_s - 1)/(2\epsilon_s + 1)\right] - 9.1 \qquad (4.5)$$

where: σ = surface tension in ergs/cm^2

ϵ_s = static dielectric constant

Equation (4.5) is good for all liquids with zero dipole moment. These include: hexane, octane, benzene, p-xylene, carbon tetrachloride, p-dichlorobenzene, CS_2 , H_2 , N_2 , O_2 , A , Br_2 , Cl_2 .

For molecules with non-zero dipole moments, the relation is of similar form, but includes the index of refraction, η , instead of the dielectric constant. This correlation is shown below:

$$\sigma = 286.0\left[(\eta^2-1)/(2\eta^2+1)\right] - 28.6 \quad (4.6)$$

The previous correlations between surface tension and other physical properties (viscosity, density, dielectric constant, index of refraction) are very useful, since surface tension data is difficult to find at more than one temperature.

The previous few paragraphs have been an introduction to the concept of surface tensions. For a more detailed discussion on this topic, references (23) to (28) are recommended.

Chapter 5

Results

5.1 Analytical Results

Analytical results have been obtained using the energy balance method of Bell (5). The results include fast neutrons in the test fluid and fission fragments in the test fluid.

For the case of fast neutrons in the test fluid, the initial energy of the PKOA was 2.12 Mev. This initial energy value is listed on the graphs as "EPKA" (energy of the primary knock-on atom). (The value of 2.12 Mev for this initial energy was obtained through an analysis by Bell((5) pg. 173) considering the Pu-Be neutron source and its associated energy spectrum.)

The test fluids for which analytical results were obtained are:

1.) Water (FIG.5.1 and FIG.5.2)
2.) Propylene Glycol (FIG.5.3 and FIG.5.4)
3.) Benzene (FIG.5.5 and FIG.5.6)
4.) Acetone (FIG.5.7 and FIG.5.8.)
5.) Nitromethane (FIG. 5.9 and FIG.5.10)

The results are given in the form of a graph of the threshold superheat (amount of superheat) versus the system pressure, for a number of "a" values.

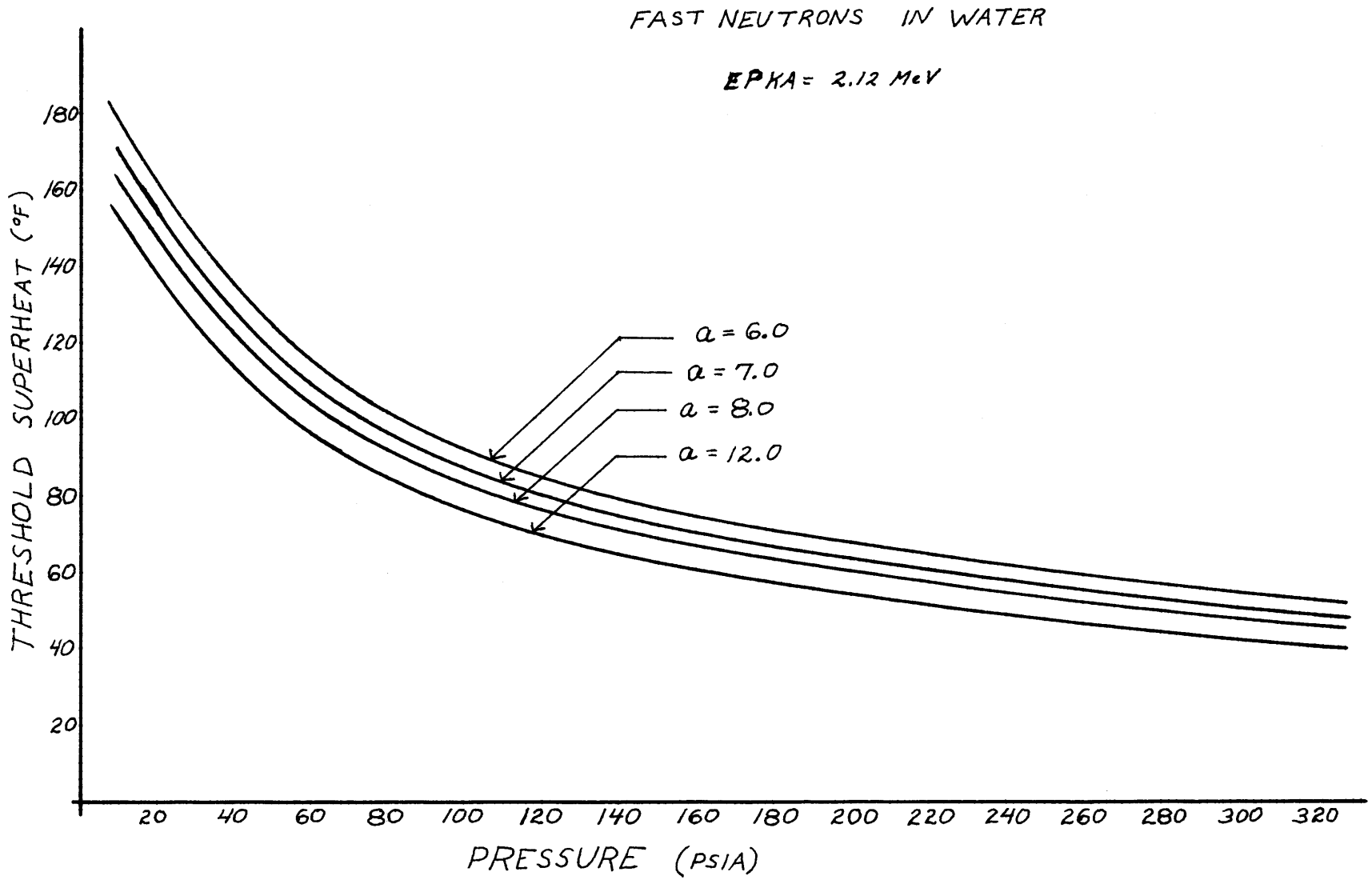

FAST NEUTRONS IN WATER
EPKA = 2.12 MeV
THRESHOLD SUPERHEAT (°F)
180
160
140
120
100
80
60
40
20
a = 6.0
a = 7.0
a = 8.0
a = 12.0
20 40 60 80 100 120 140 160 180 200 220 240 260 280 300 320
PRESSURE (PSIA)

FIG. 5.1.

FISSION FRAGMENTS IN WATER

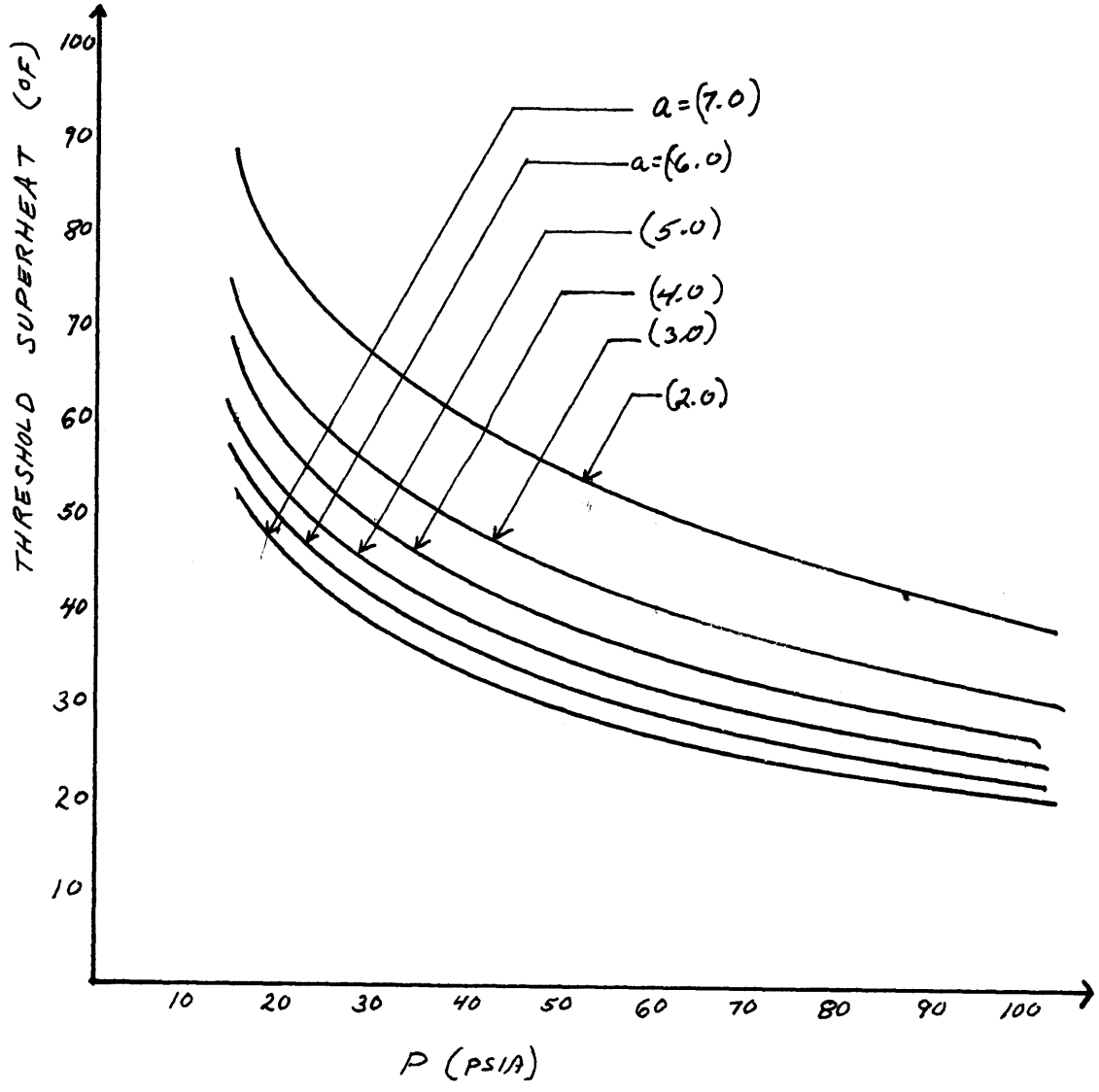


FIG 5.2

FAST NEUTRONS IN PROPYLENE GLYCOL

EPKA = 2.12 MeV

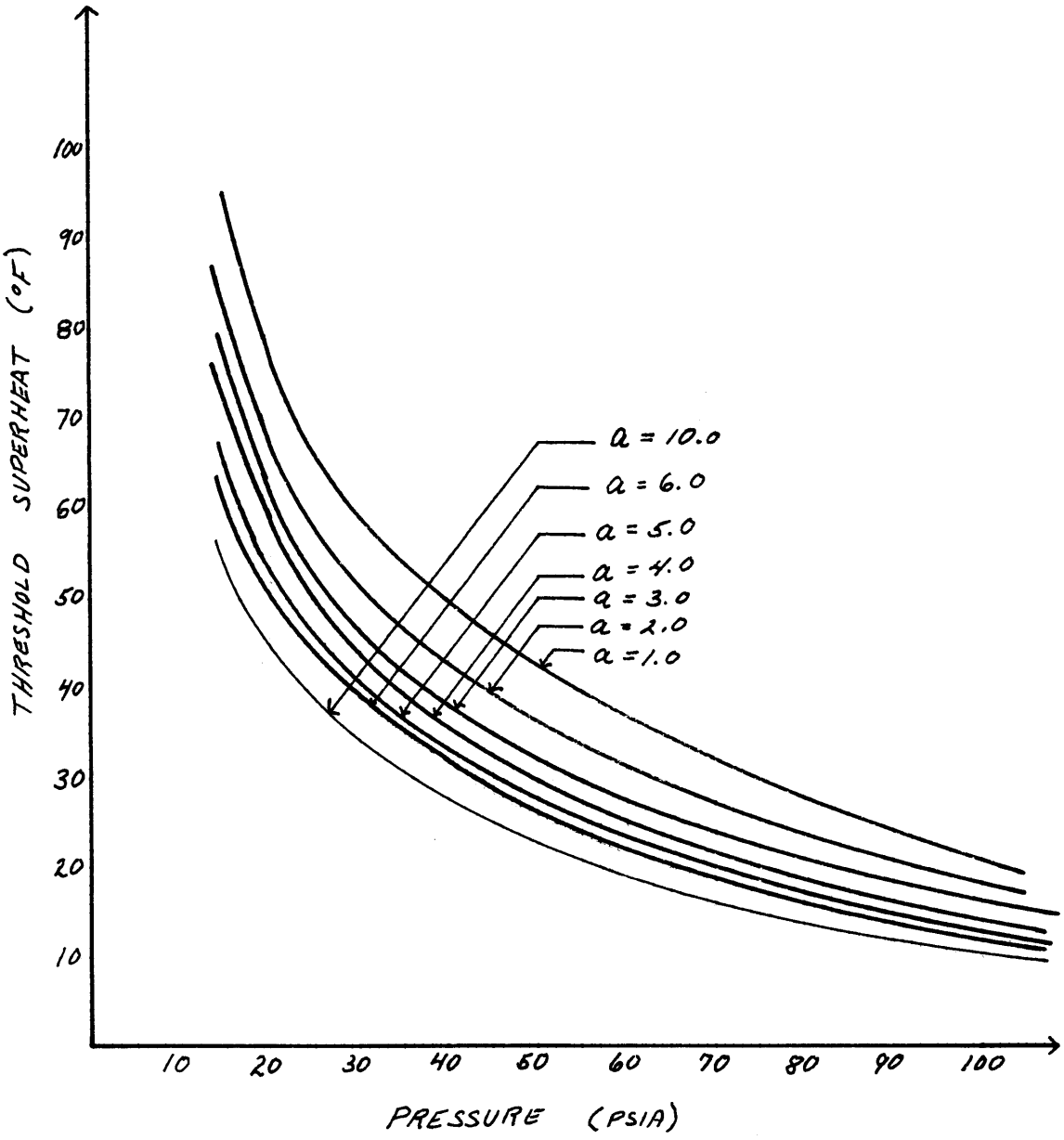


FIG. 5.3

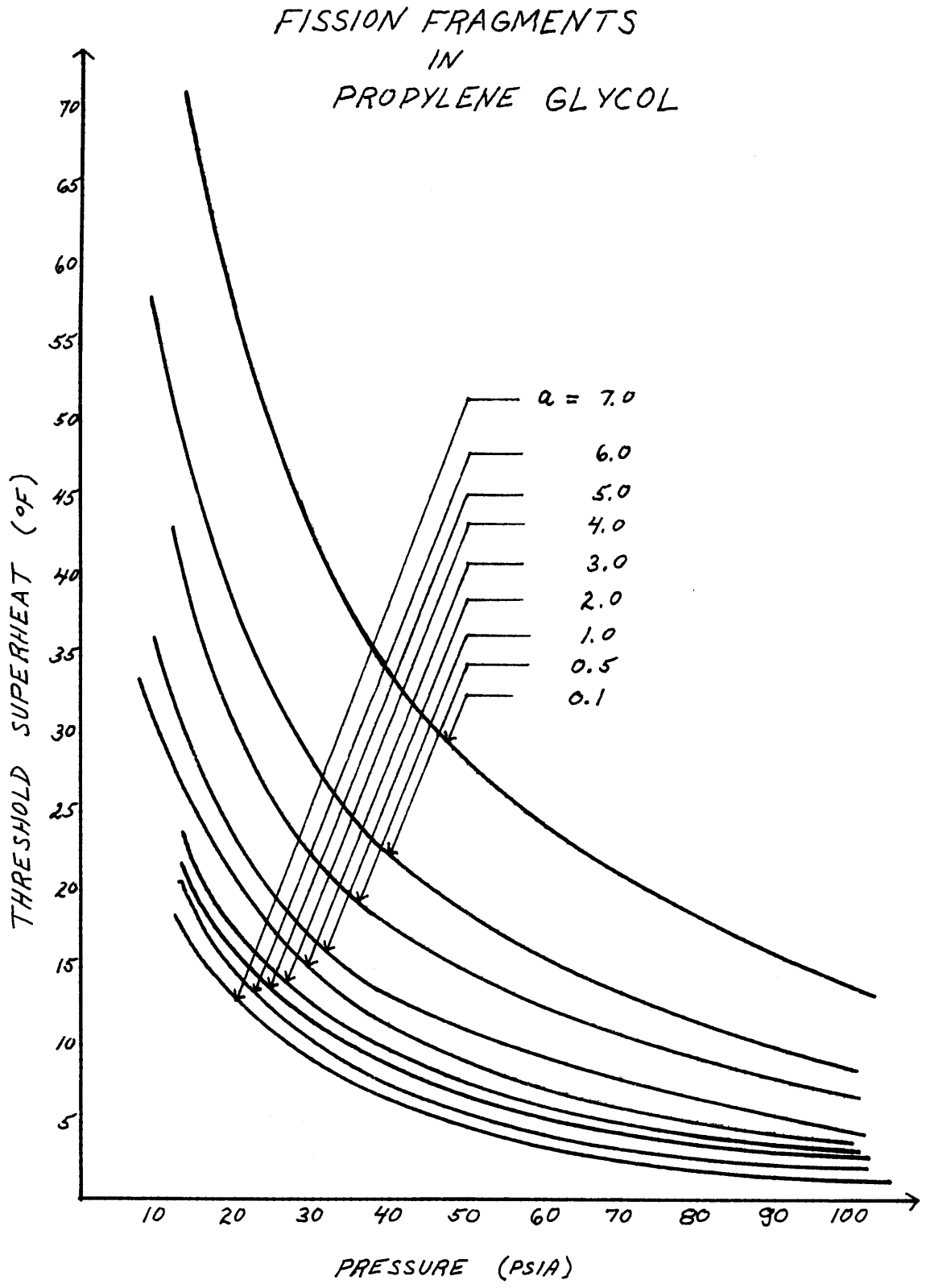

FISSION FRAGMENTS
IN
PROPYLENE GLYCOL
THRESHOLD SUPERHEAT (°F)
70
65
60
55
50
45
40
35
30
25
20
15
10
5
a = 7.0
6.0
5.0
4.0
3.0
2.0
1.0
0.5
0.1
10
20
30
40
50
60
70
80
90
100
PRESSURE (PSIA)

FIG. 5.4

FAST NEUTRONS IN BENZENE

EPKA = 2.12 MeV

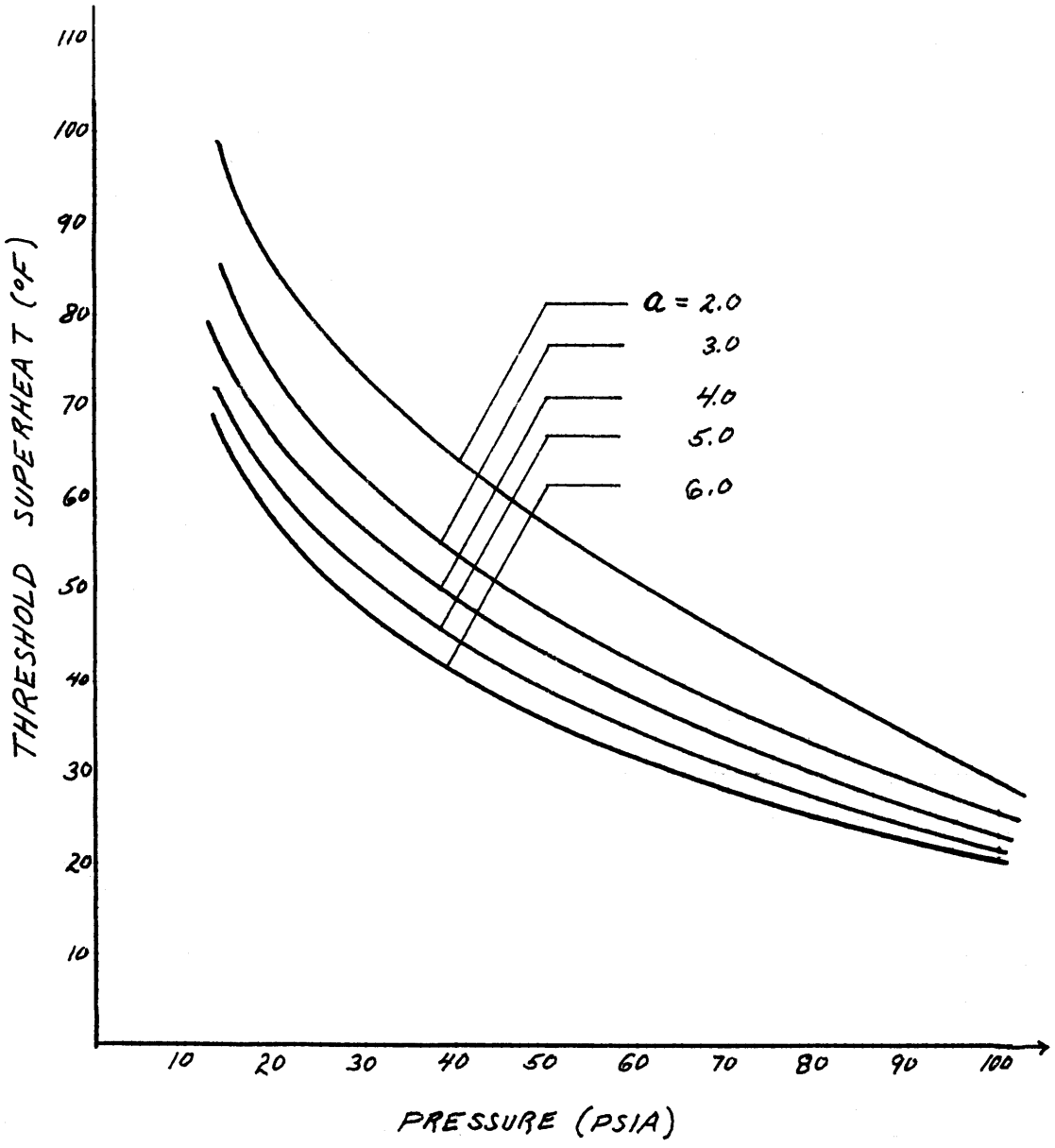


FIG. 5.5

FISSION FRAGMENTS
IN
BENZENE

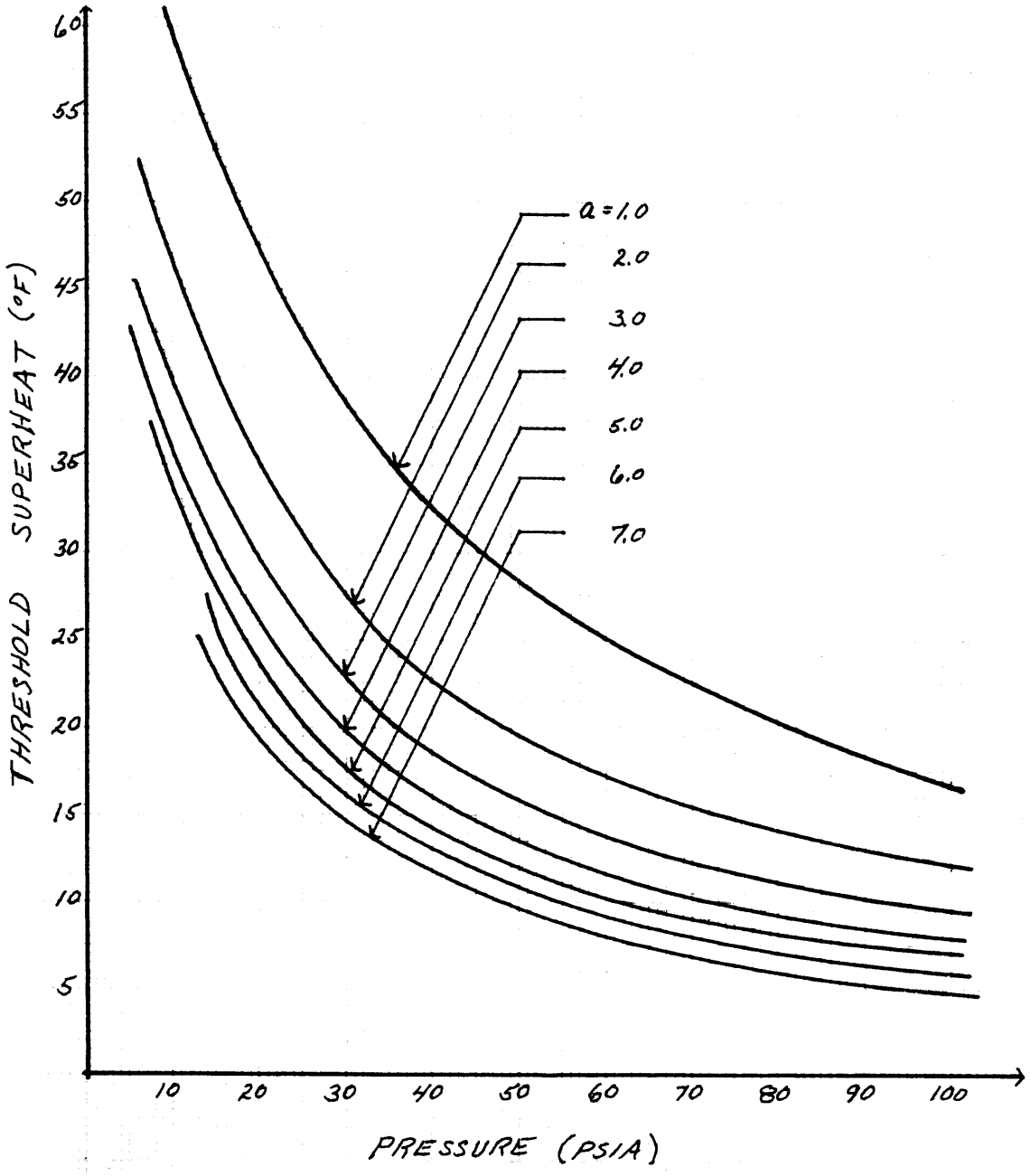


FIG. 5.6

FAST NEUTRONS
IN
ACETONE
EPKA = 2.12 MeV

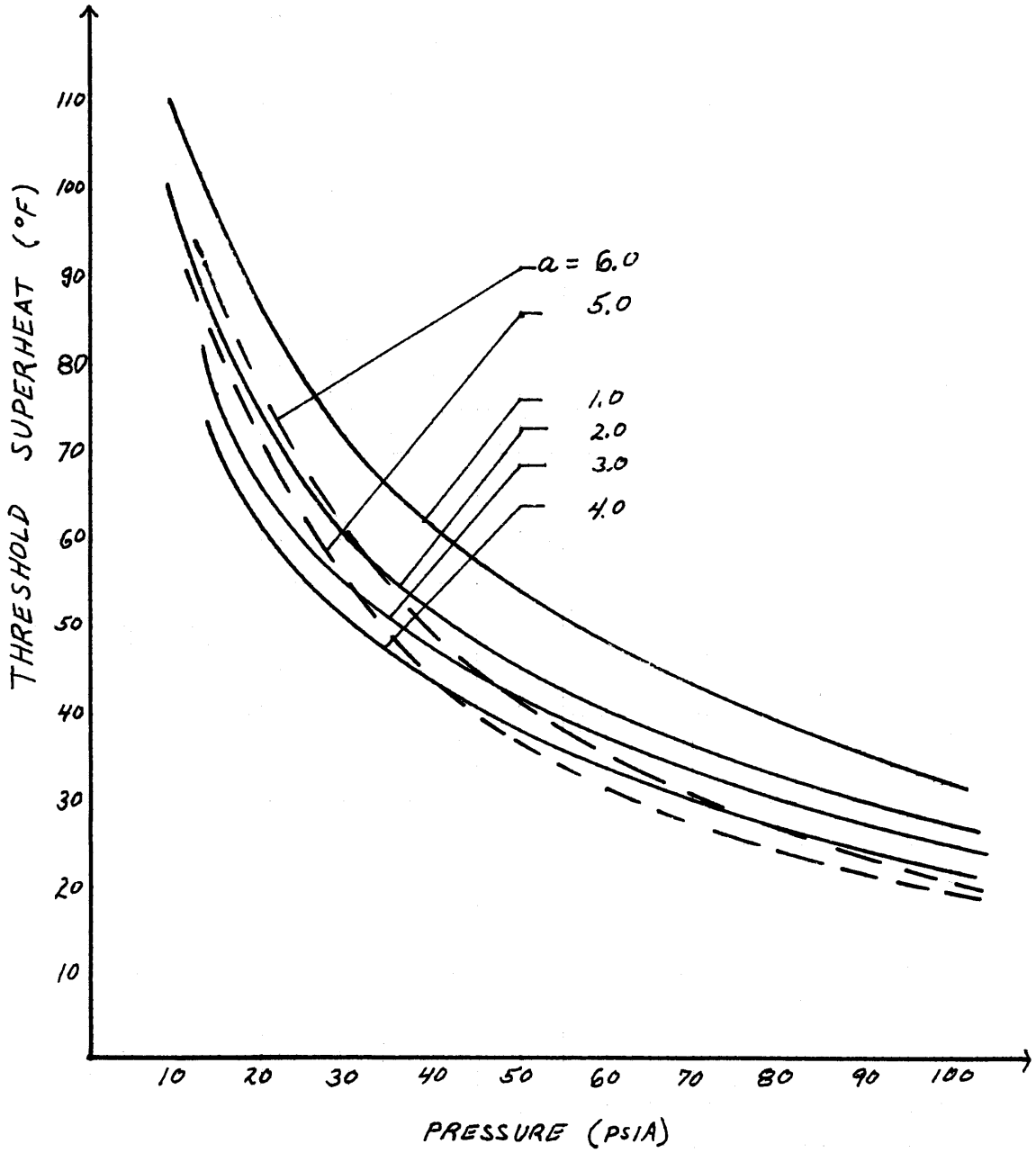


FIG. 5.7.

FISSION FRAGMENTS
IN
ACETONE

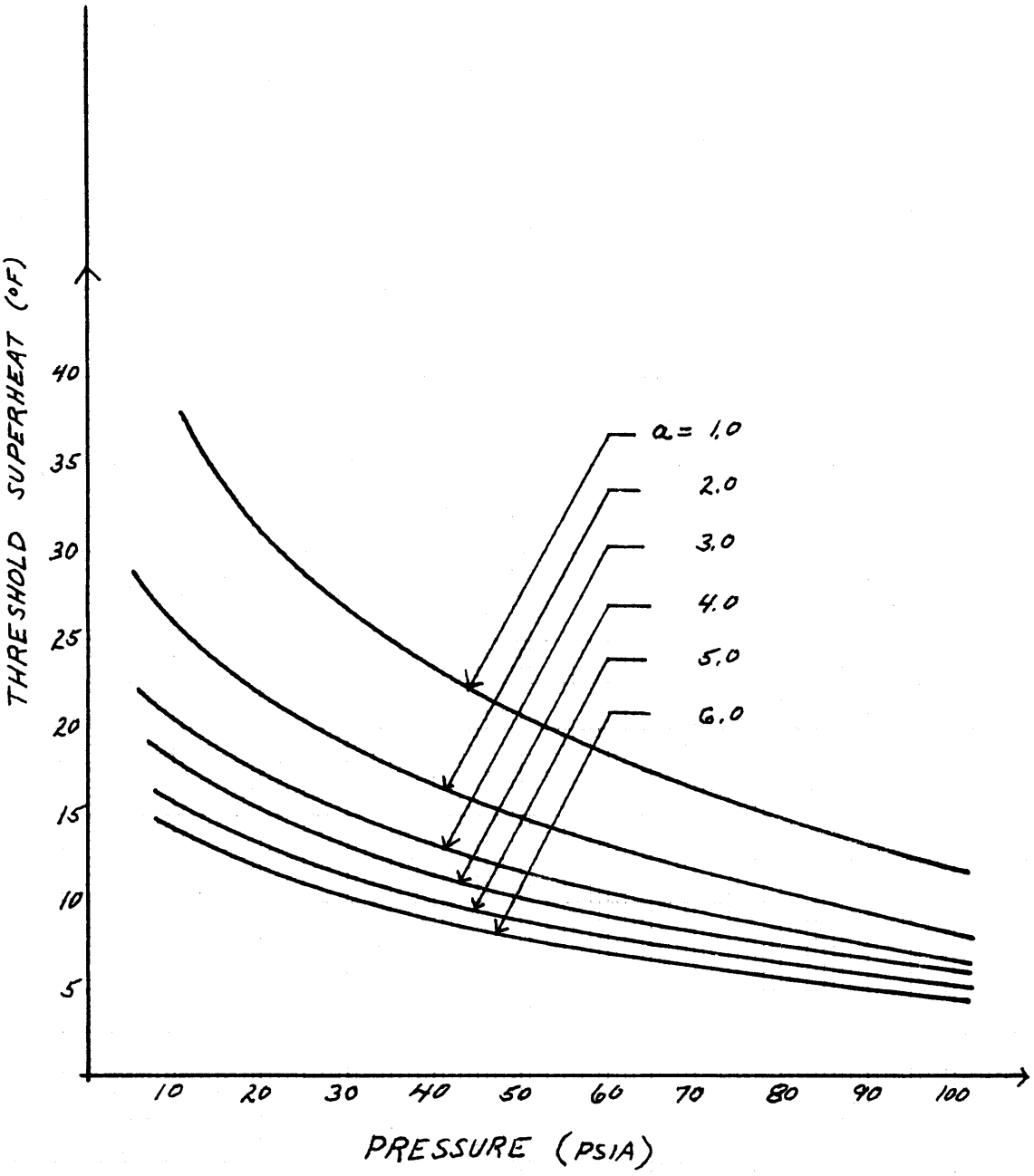


FIG. 5.8

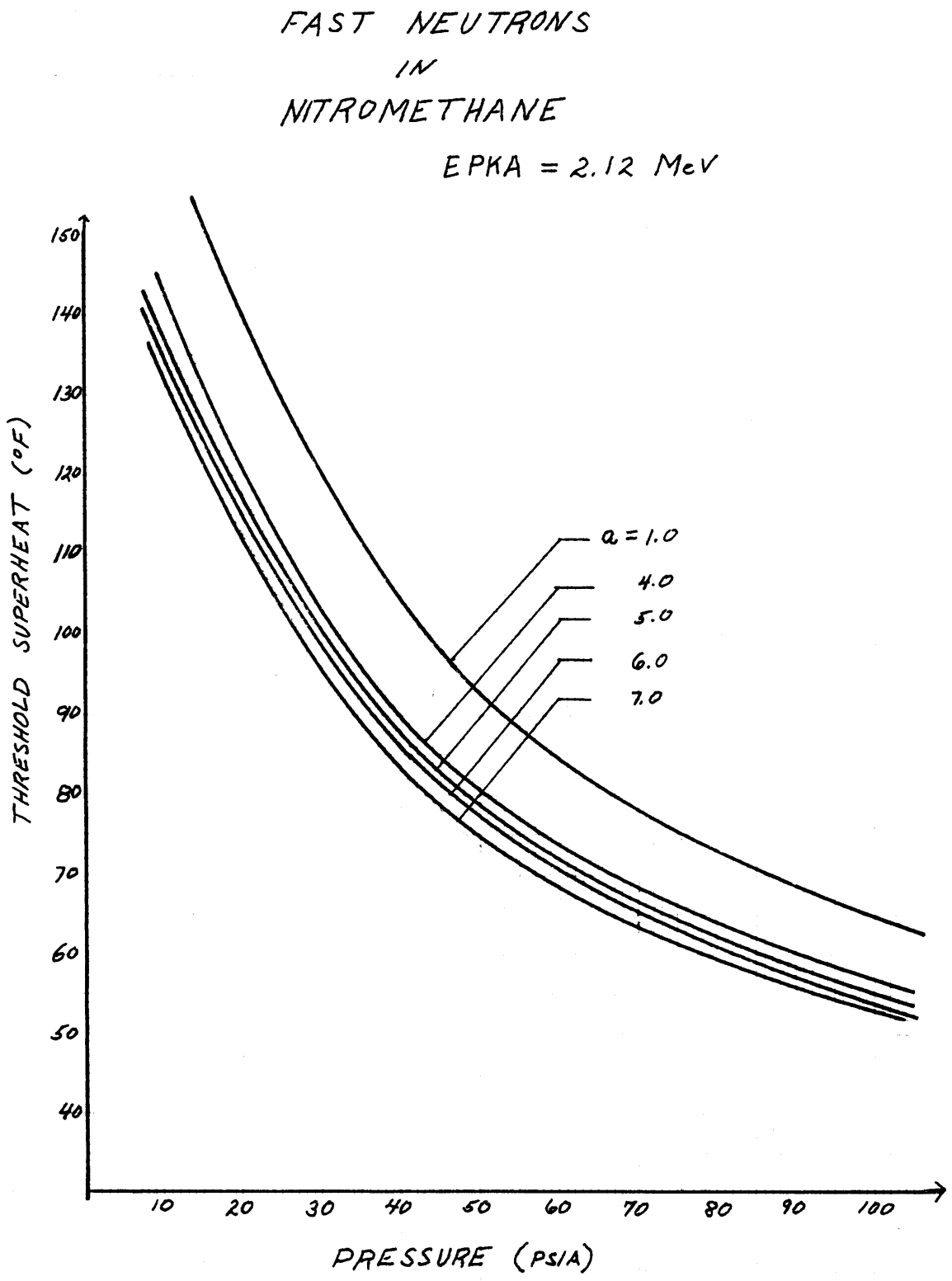

FAST NEUTRONS
IN
NITROMETHANE
EPKA = 2.12 MeV
THRESHOLD SUPERHEAT (°F)
150
140
130
120
110
100
90
80
70
60
50
40
a = 1.0
4.0
5.0
6.0
7.0
10
20
30
40
50
60
70
80
90
100
PRESSURE (PSIA)

FIG. 5.9

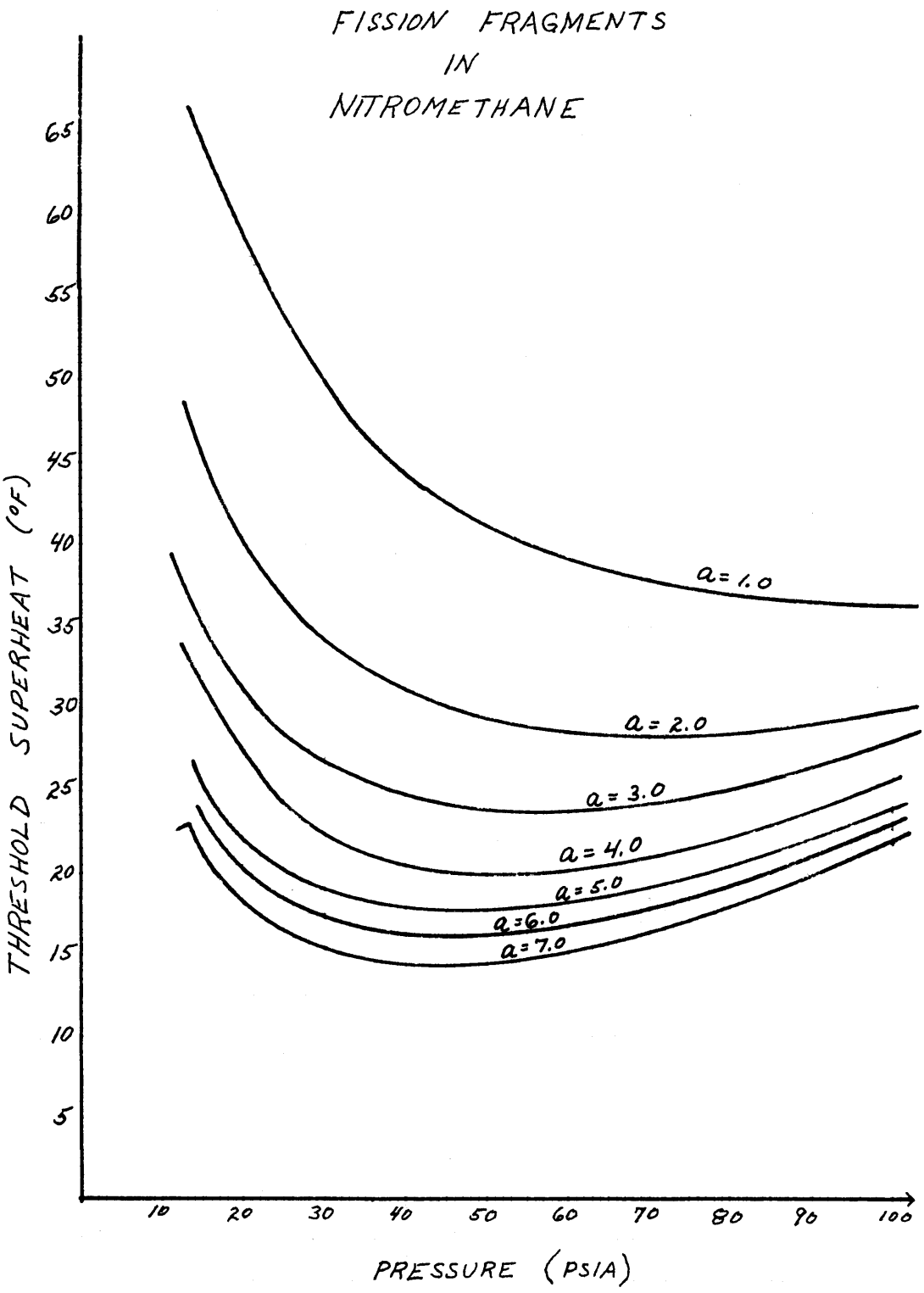

FISSION FRAGMENTS
IN
NITROMETHANE
THRESHOLD SUPERHEAT (°F)
65
60
55
50
45
40
35
30
25
20
15
10
5
a = 1.0
a = 2.0
a = 3.0
a = 4.0
a = 5.0
a = 6.0
a = 7.0
10
20
30
40
50
60
70
80
90
100
PRESSURE (PSIA)

FIG. 5.10

5.2 Experimental Results

Experimental results were obtained for fission fragments in water and propylene glycol.

For water, fission fragment data was obtained at four pressures: 14.7psia, 32.7psia, 53.7psia, and 74.2psia . These were the same pressures as were tested by Tso(6), since one of the objectives of this work was to attempt to confirm the results obtained by Tso for the fission fragment case.

The results obtained in this work are shown on FIG.5.11. These results and the results obtained by Tso are listed together in TABLE 5.1, shown below.

TABLE 5.1

PRESSURE (psia)	AMOUNT SUPERHEAT (°F)	
	TSO (6)	OBERLE
14.7	63.5 ± 2	62.4 ± 2.5
32.7	49.3 ± 3	48.5 ± 2.0
53.7	38.7 ± 2.5	39.2 ± 4.1
74.2	34.2 ± 2.4	33.3 ± 2.*

As is seen above, the results obtained in this work agree quite well with those obtained by Tso (6).

FISSION FRAGMENTS IN WATER
EXPERIMENTAL RESULTS

OBERLE: I

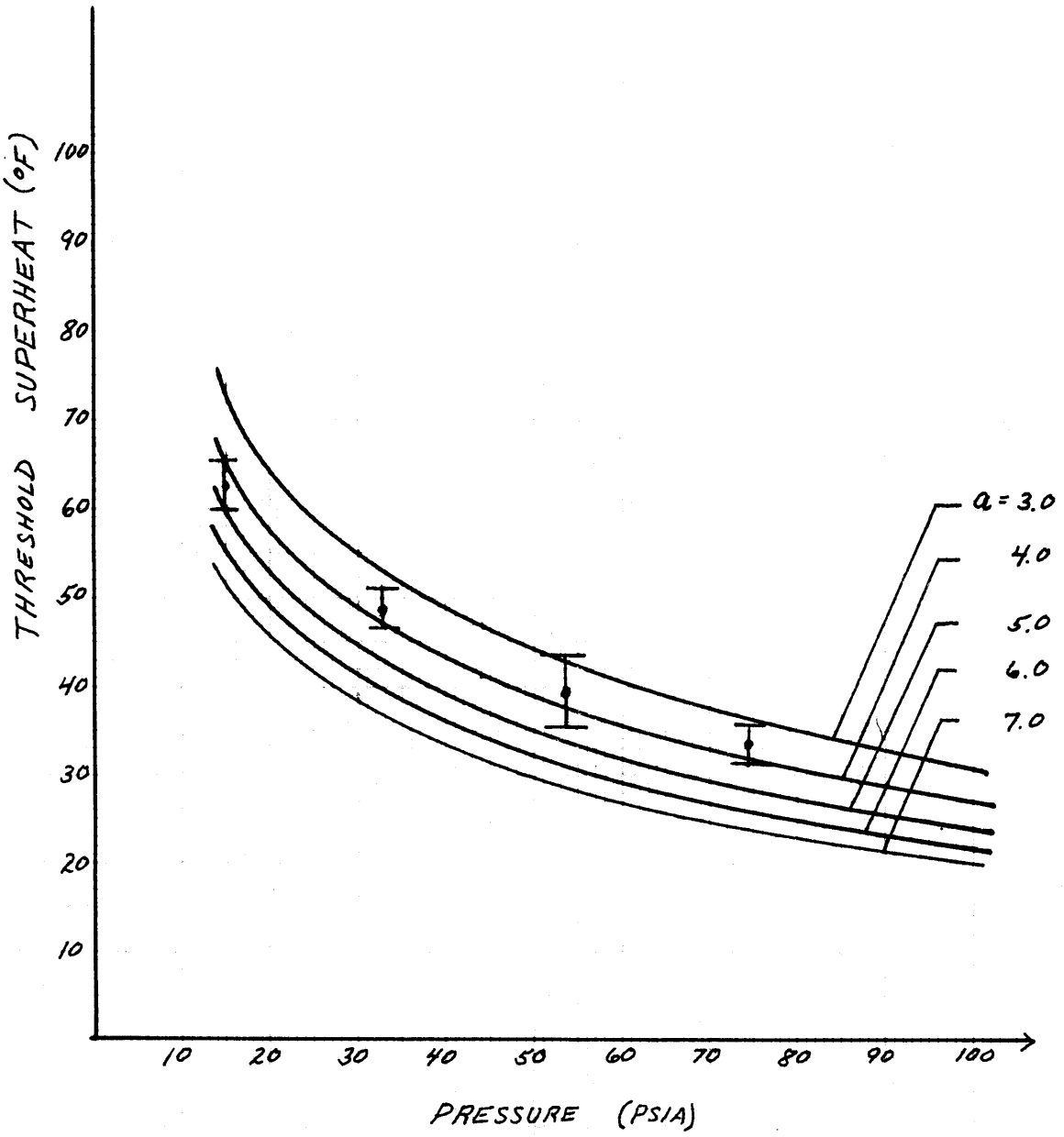


FIG. 5.11

For the case of fast neutrons , only one data point was obtained. This was for the case of fast neutrons in water at 55 psia. A large number of experimental runs were required to obtain one good result for this case. This was due mainly to the fact that this data was at high temperatures , and the convection generator was not very effective in keeping the bubble centered. Therefore, due to the difficult and time consuming experimentation involved, further work was not done with fast neutrons, since work was still to be done with organic liquids.

The result obtained for the single fast neutron result is shown in TABLE 5.2, along with the results of Tso (6) at this same pressure.

TABLE 5.2

	PRESSURE psia	AMOUNT SUPERHEAT (°F)	TEMPERATURE RAMP (°F/min)
OBERLE	55	105.0	2.0
TSO(6)	55	102.4	1.6
"	55	105.9	1.35

Bell (5) and Tso (6) also encountered considerable difficulties with the fast neutrons in water. Tso , ((6) pg. 68) has mentioned the difficulties and reliability of their work.

The experimental data obtained for propylene glycol exposed to fission fragments is shown in FIG.5.12.

The superheat data was obtained at four pressures: 14.7psia, 20psia, 40psia, 60psia.

Considerable difficulty was encountered for the higher pressure (40psia and 60 psia) since high temperatures (around 500°F) were required, and the convection generator was not able to keep the bubble centered. Due to the convection generator problem, more than one bubble was placed into the test chamber for each experimental run, so that if one or more bubbles drifted out of sight, there would, it was hoped, still remain at least one test bubble in sight. This was not always the case, since very often all of the test bubbles drifted out of sight. There were not as many good experimental runs for these pressures as there were for the two lower pressures studied. Due to this fact, and due to the presence and movement of a large number of test bubbles per experimental run, the data obtained at these higher pressures is not considered to be as accurate as that for the lower pressures.

5.3 Sensitivity Study of Superheat Data

The minimum superheat conditions are especially sensitive to the surface tension of the fluid and to the energy deposition of the charged particle. It was

FISSION FRAGMENTS IN
PROPYLENE GLYCOL
EXPERIMENTAL RESULTS

OBERLE: I

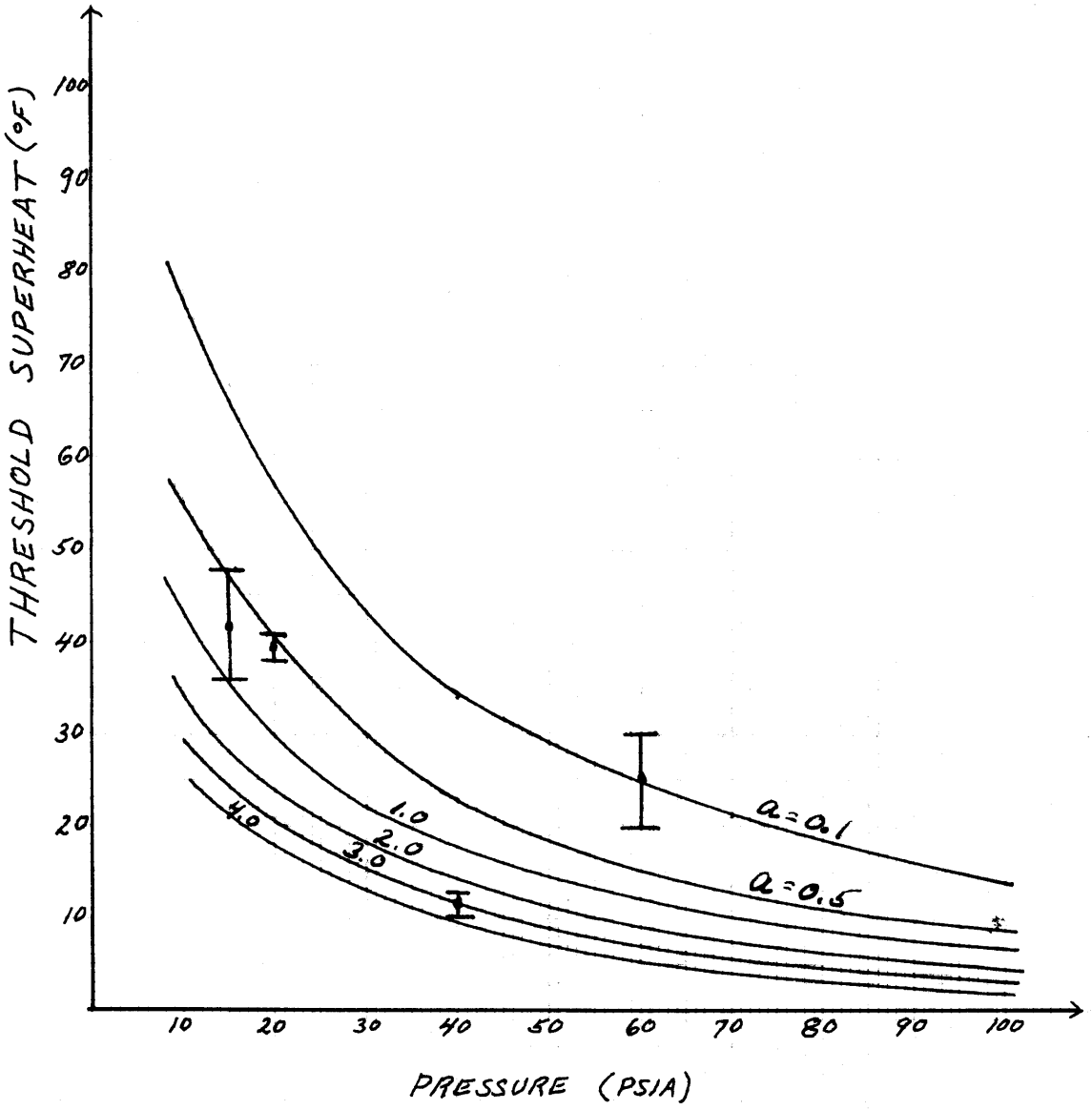


FIG. 5.12

reported by Connolly (33) that variations in these two factors significantly changed the analytical results of his group's work.

Therefore, a sensitivity study was made for water and propylene glycol exposed to fission fragments.

For the case of water, a ± 7% variation in the surface tension was studied. The result on the minimum superheat is shown in FIG.5.13 .

A 20% variation in the mean charge of the radiation particle was considered. (The mean charge is the most sensitive factor in the energy deposition calculation. And, as suggested by Bell (5), a 20% variation is conceivable.) The effect of this large uncertainty is shown in FIG.5.14 .

It was seen that a positive uncertainty in the surface tension increased the minimum superheat, while a negative uncertainty decreased the minimum superheat. In the case of the mean charge a positive uncertainty decreased the mean superheat while a negative uncertainty increased it. Therefore, when these two uncertainties were combined to obtain a maximum uncertainty in the minimum superheat, they were combined in the following manner:

maximum positive uncertainty---	+7% in surface tension -20% in mean charge
maximum negative uncertainty--	-7% in surface tension +20% in mean charge

The result of the maximum uncertainties are shown in FIG.5.15 .

FISSION FRAGMENTS IN WATER

$\pm 7\%$ UNCERTAINTY IN SURFACE TENSION

I OBERLE DATA

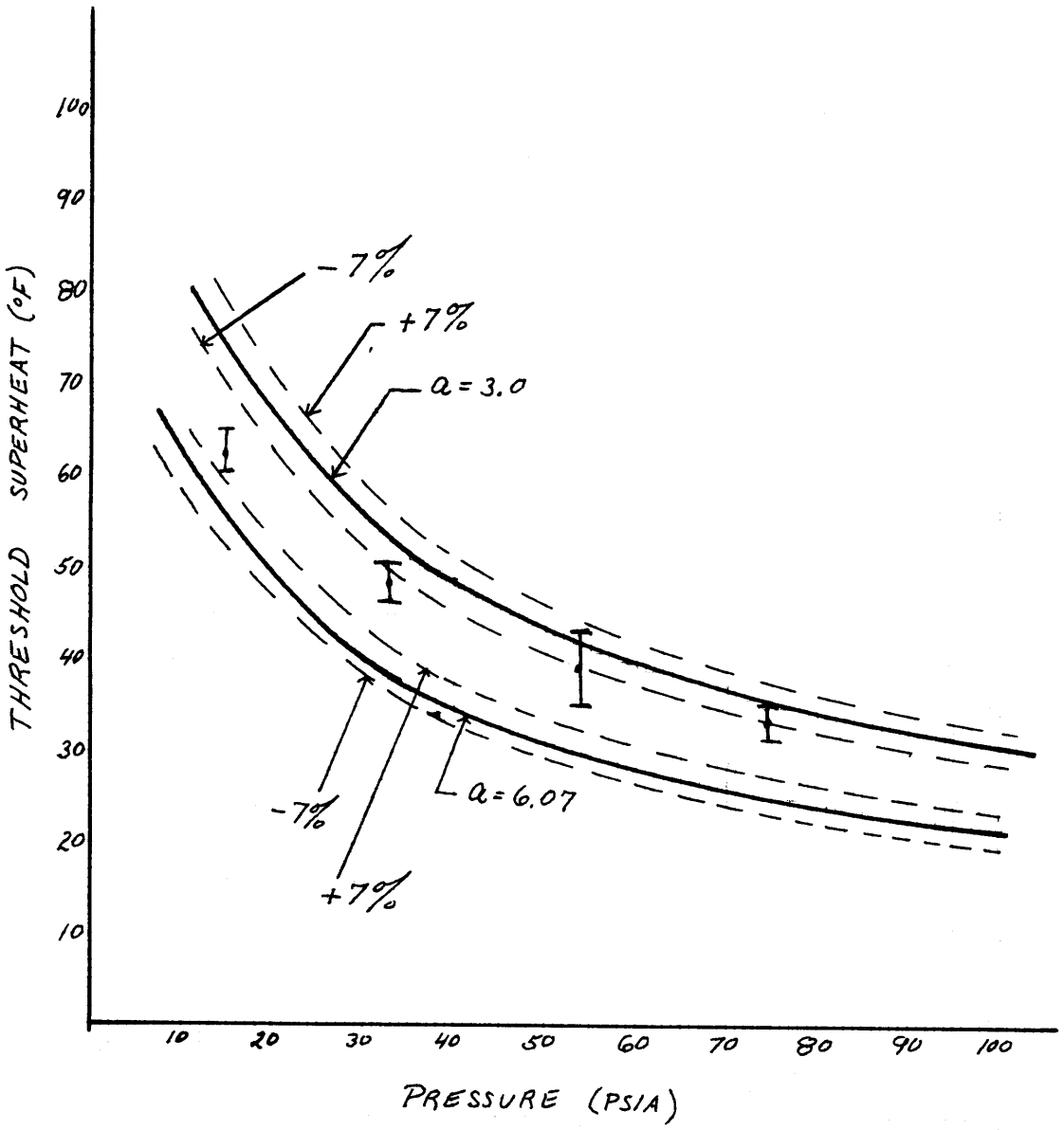


FIG. 5.13

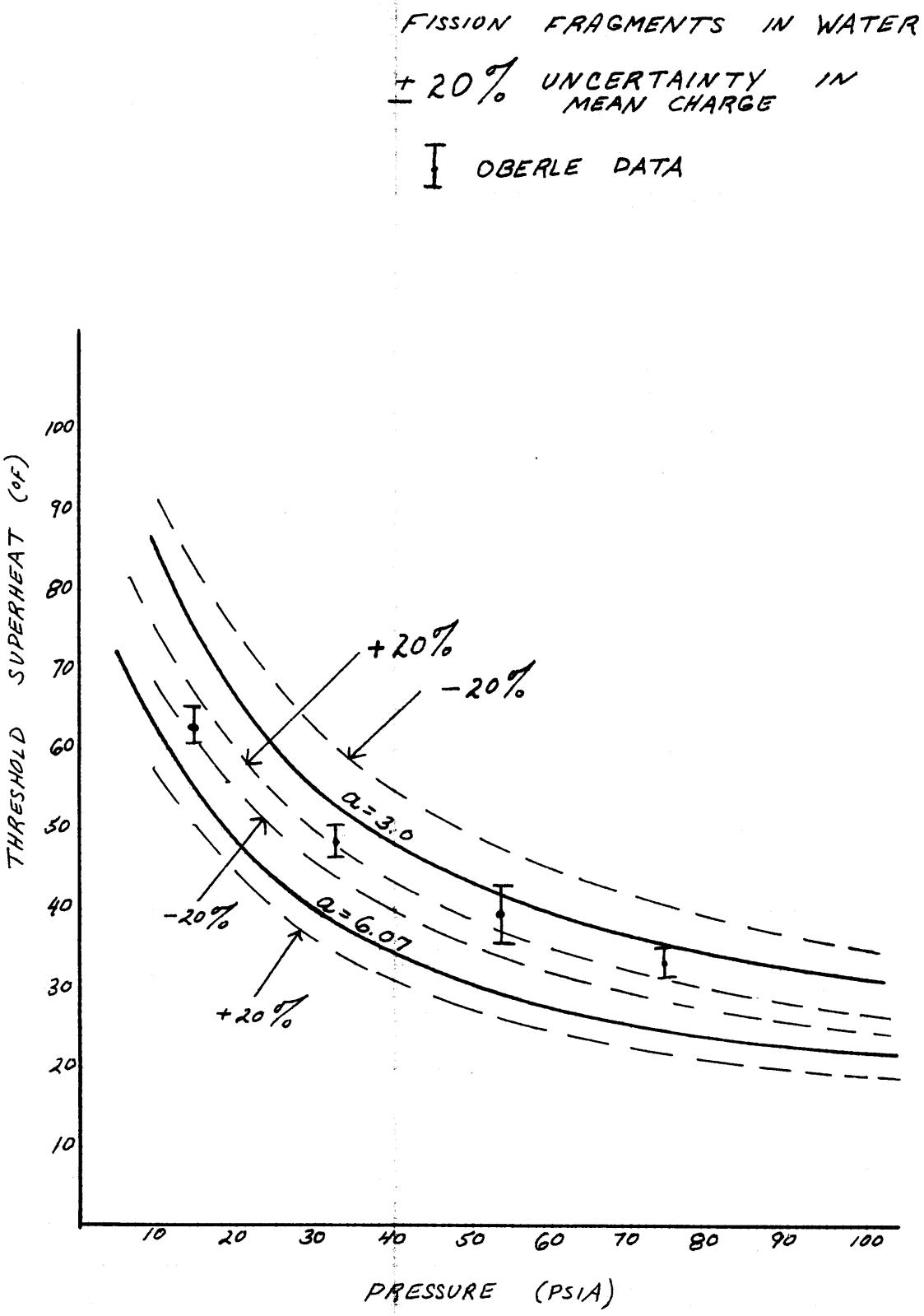

FISSION FRAGMENTS IN WATER
± 20% UNCERTAINTY IN MEAN CHARGE
OBERLE DATA
THRESHOLD SUPERHEAT (°F)
PRESSURE (PSIA)
+20%
-20%
a=3.0
a=6.07
-20%
+20%
10
20
30
40
50
60
70
80
90
100

FIG. 5.14

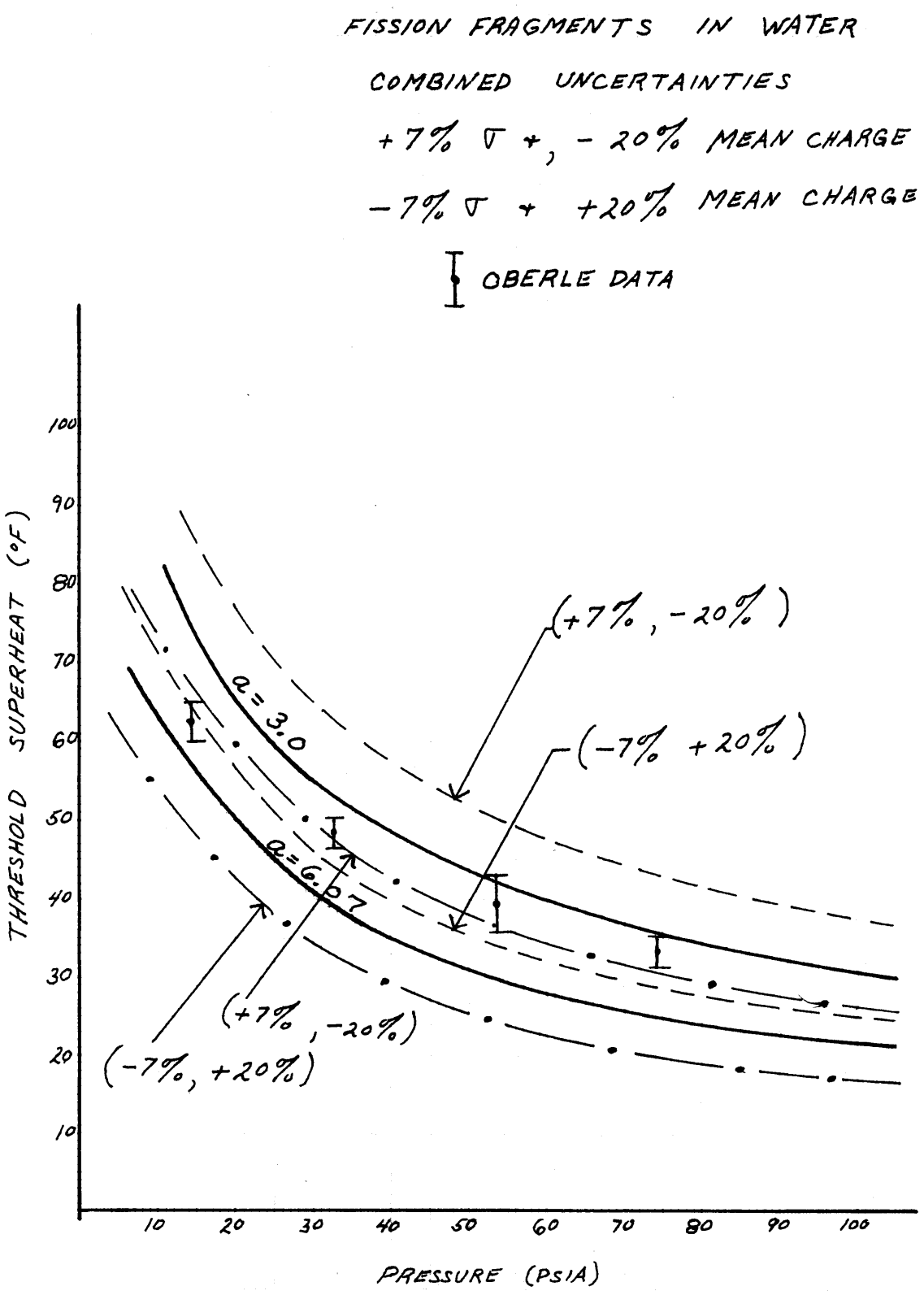

FISSION FRAGMENTS IN WATER
COMBINED UNCERTAINTIES
+7% σ +, - 20% MEAN CHARGE
-7% σ + +20% MEAN CHARGE
OBERLE DATA
THRESHOLD SUPERHEAT (°F)
PRESSURE (PSIA)
100
90
80
70
60
50
40
30
20
10
10
20
30
40
50
60
70
80
90
100
α = 3.0
α = 6.07
(+7%, -20%)
(-7% +20%)
(+7%, -20%)
(-7%, +20%)

FIG 5.15

This same type of sensitivity study was made for the case of propylene glycol. The percent variations used were:

maximum uncertainties in surface tension-- ±5%

maximum uncertainties in mean charge -- ± 20%

The directional affect of each uncertainty is the same as for water. The results of these studies are shown in FIG.5.16, FIG.5.17, FIG.5.18 .

5.4 Comparison with other results

Experimental results have been obtained by other researchers.

Deitrich (3) has obtained data for the case of fission products in water. His results are shown in FIG.5.19, along with the results from this work.

El-Nagdy (4) has obtained experimental data for Benzene and Acetone exposed to fast neutrons at atmospheric pressure. The fast neutrons were at different energies than those from the Pu-Be source used in this work. His experimental data is shown with the analytical results from the energy balance method of Bell(5). (Adjustments had been made to conform with the neutron energies used by El-Nagdy.) The results are shown in FIG.5.20 and FIG.5.21. (The 14.1 Mev neutrons correspond to an EPKA of 4.0 , and the 2.45 Mev neutrons correspond to an EPKA of 0.695 Mev.

FISSION FRAGMENTS IN
PROPYLENE GLYCOL
± 5% UNCERTAINTY IN
SURFACE TENSION

I EXPERIMENTAL DATA (OBERLE)

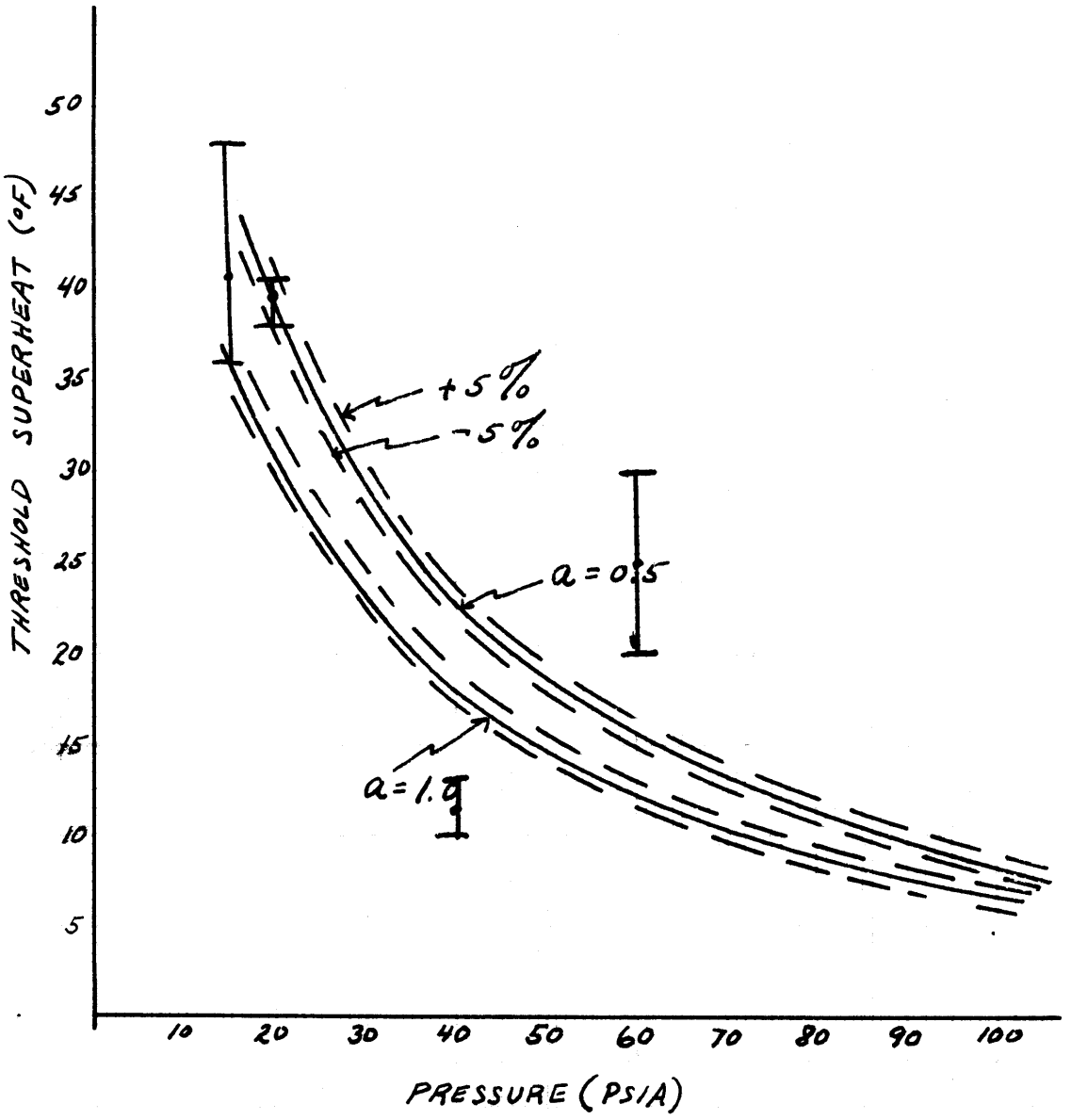


FIG. 5.16

FISSION FRAGMENTS IN PROPYLENE GLYCOL

$\pm 20\%$ UNCERTAINTY IN MEAN CHARGE

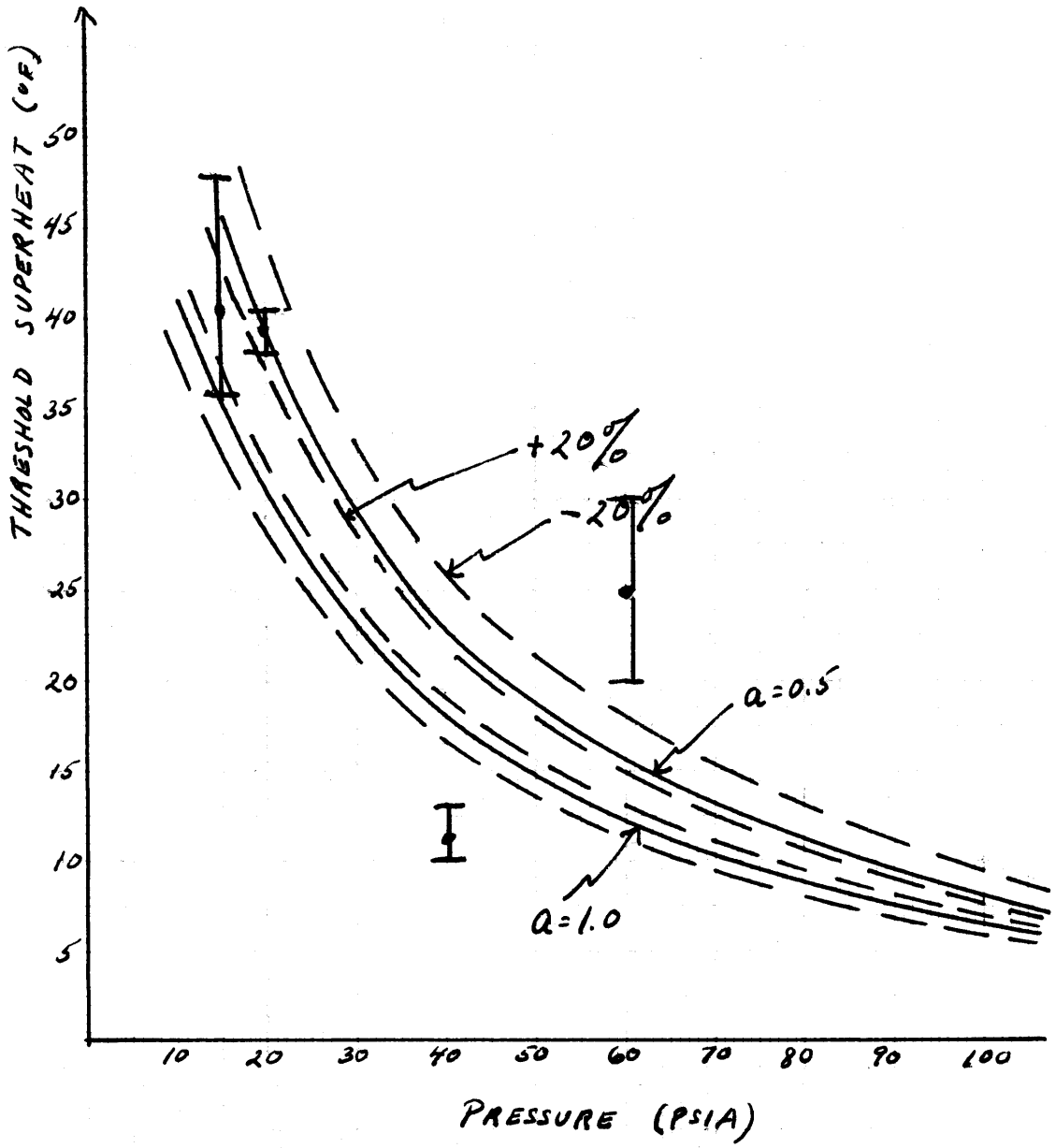


FIG. 5.17

FISSION FRAGMENTS IN PROPYLENE GLYCOL

COMBINED UNCERTAINTIES

+5%, −20%

−5%, +20%

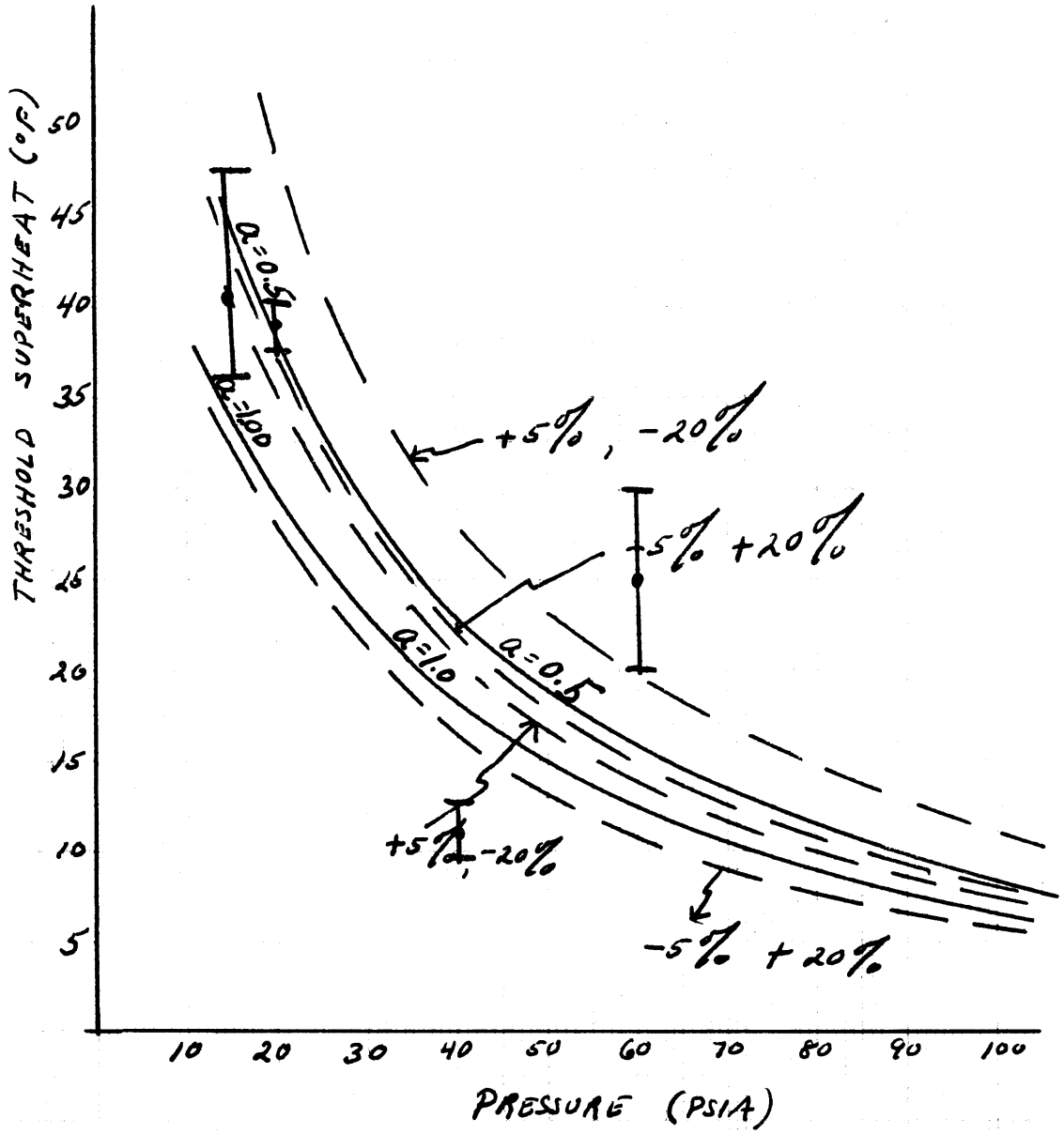


FIG 5.18

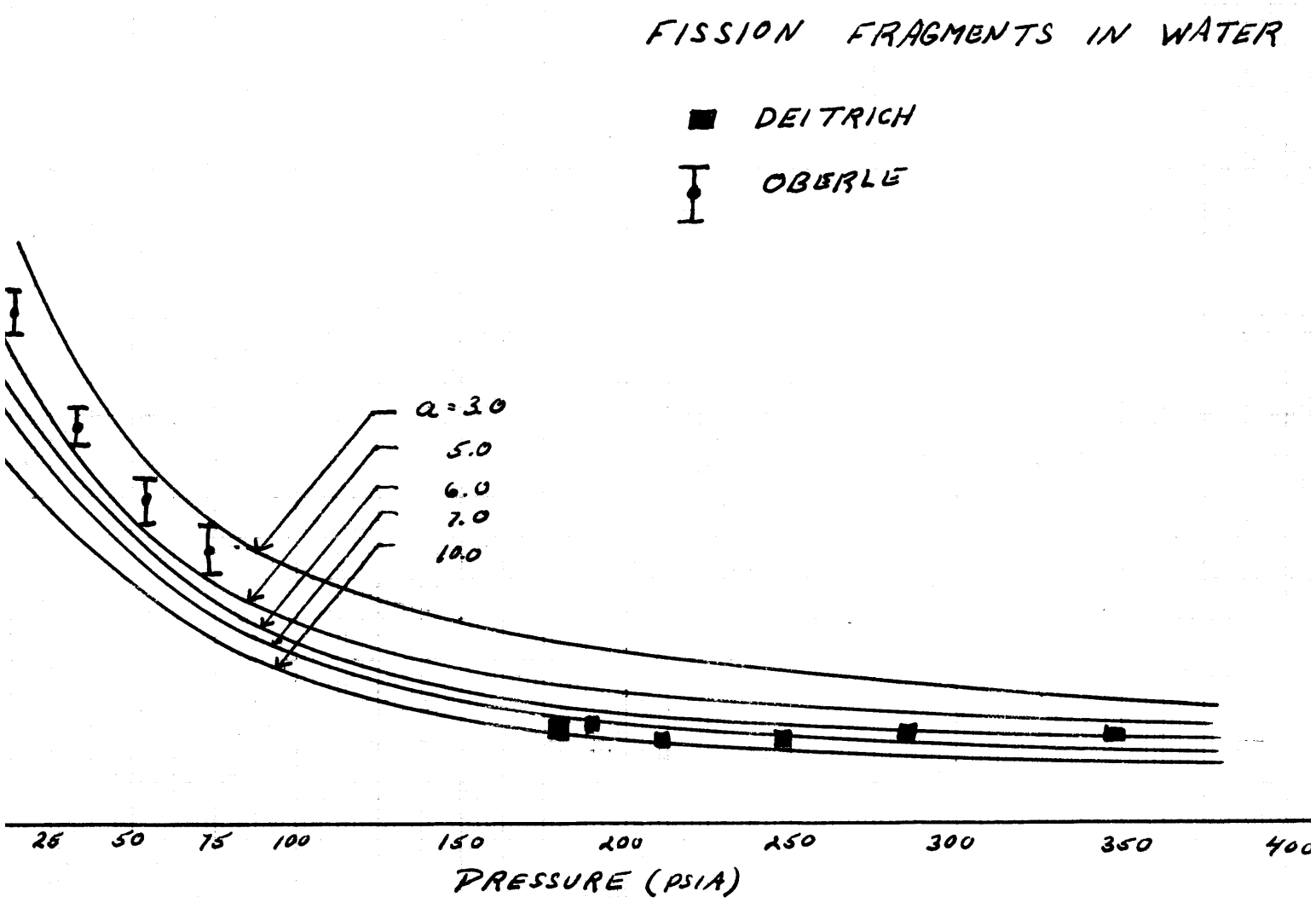

FISSION FRAGMENTS IN WATER
DEITRICH
OBERLE
a = 3.0
5.0
6.0
7.0
10.0
25
50
75
100
150
200
250
300
350
400
PRESSURE (PSIA)

FIG. 5.19

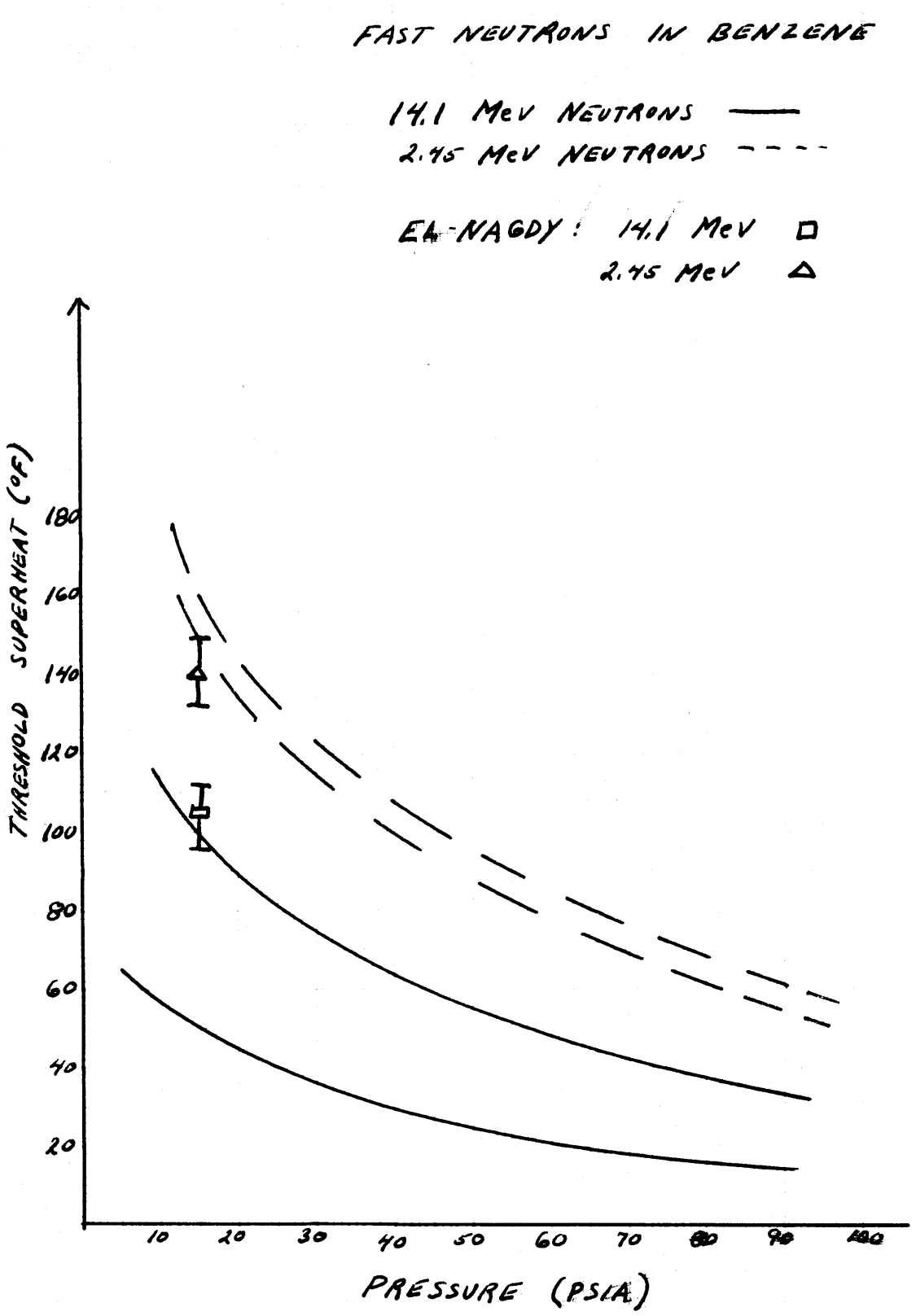

FAST NEUTRONS IN BENZENE
14.1 MeV NEUTRONS
2.45 MeV NEUTRONS
EL-NAGDY: 14.1 MeV
2.45 MeV
THRESHOLD SUPERHEAT (°F)
180
160
140
120
100
80
60
40
20
10
20
30
40
50
60
70
80
90
100
PRESSURE (PSIA)

FIG. 5.20

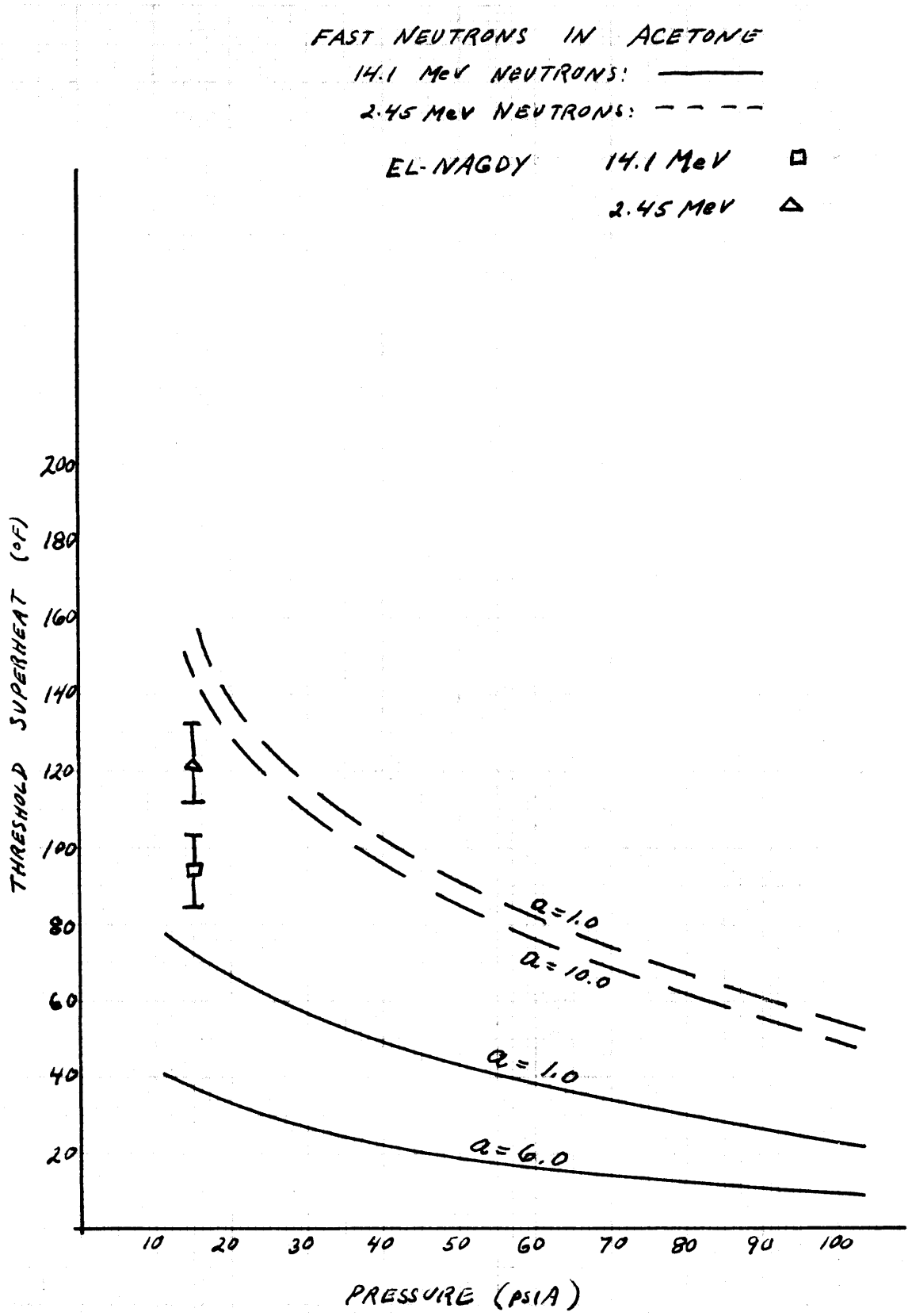

FAST NEUTRONS IN ACETONE
14.1 MeV NEUTRONS:
2.45 MeV NEUTRONS:
EL-NAGDY 14.1 MeV
2.45 MeV
THRESHOLD SUPERHEAT (°F)
200
180
160
140
120
100
80
60
40
20
a=1.0
a=10.0
a=1.0
a=6.0
10
20
30
40
50
60
70
80
90
100
PRESSURE (psiA)

FIG. 5.21

Chapter 6

Conclusions and Recommendations

6.1 Conclusions

The experimental results for fission fragments in water have been shown in FIG.5.11. These results can be considered very reliable, since the nucleation event was very noticeable, and, more significantly, this data was very close to the data of Tso(6), who also did experimental work under these conditions. It is therefore reasonable to conclude that these minimum superheat results for water exposed to fission fragments, can be considered very accurate and reliable.

These experimental results fall between "a" values of approximately 3.5 and 4.5. In any case, the "a" value is less than 6.07,as predicted by Bell (5).

The experimental results of Deitrich(3) have been plotted over calculated "a" curves. (FIG.5.19) These results varied from an "a" value of 10.0, to an "a" value of 5.0 Deitrich's data also seems to be fairly reliable, with the uncertainties in his superheat data of $\pm$ 0.5 F .

Therefore, the experimental evidence seems to point to the conclusion that the "constant a " theory of Bell (5) does not hold for the case of fission fragments in water.

There exists, of course, the possibility that there is some error or uncertainty in the analytical calculations for the constant "a" curves.

A sensitivity study was made on the parameters which have the greatest affect on the superheat limits, and thus on the "a" curves. A $\pm$ 7% variation in the surface tension , and a $\pm$ 20% variation in the mean charge was considered for water. (FIG.5.13 and FIG.5.14) These two uncertainties were first considered separately, and then jointly (FIG.5.15). In either case, the experimental data did not fall within the $a = 6.07$ band.

The same type of sensitivity study was made for propylene glycol exposed to fission fragments. The conclusions are similar to those for water. For these experiments with propylene glycol, all of the data points were below the $a = 6.07$ line. With all of the uncertainties mentioned above, there is a possibility that the data may all fall within the error band of a single "a" value. More study must be done on this before anything conclusive can be said on this. But in any case, it can be concluded that the "a" value is not 6.07 as stated by Bell (5).

The analytical results for the fast neutrons are similar to those of the fission fragments. The difference being that the fast neutrons require larger superheats.

In benzene and acetone (FIG.5.5 and FIG.5.6), the superheat curves are seen to go downward as "a" increases, and then to reverse direction and go up as "a" continues to increase. The reason for this is not understood at the present time, and further study needs to be done to explain this behavior.

6.2 Rayleigh's criteria

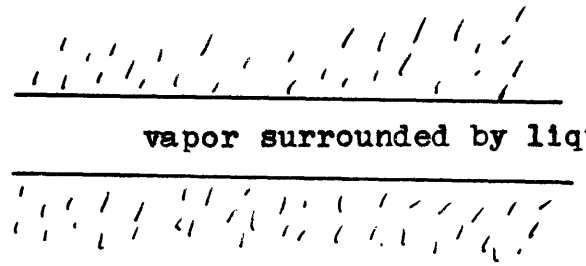
The constant "a" theory of Bell (5) concerned itself with the break up of a cylinder of vapor. It was assumed that the vapor cylinder contains disturbances of all wavelengths. These disturbances are held to be responsible for the break up of the cylinder.

Rayleigh (35) has developed a theory of jet instabilities which, in short, shows that there is a certain wavelength which will lead to the maximum rate of growth of the disturbance, and hence break up.

This certain wavelength has been found by Rayleigh for two extreme cases. In the first case, a vapor jet is surrounded by a liquid; in the second case, a liquid jet is surrounded by a vapor . A schematic of these two cases is shown in FIG.6.1 .

For the first case, that of a vapor surrounded by liquid, Rayleigh found this certain wavelength to be

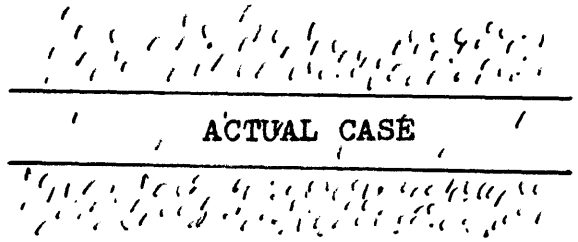
$$\lambda = (12.96)r_c \qquad (6.1)$$

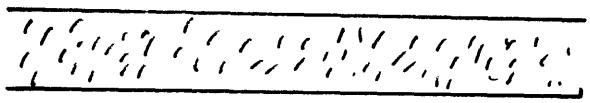


vapor surrounded by liquid

$\lambda = (12.96)r_c$

$a = 6.07$





liquid surrounded by vapor

$\lambda = (9.02)r_c$

$a = 4.79$

FIG 6.1

where λ is the wavelength

r_c is the radius of the cylinder.

For the case of a liquid jet surrounded by vapor, the wavelength is given by:

$$\lambda = (9.02)r_c$$

This wavelength is related to Bell's(5) work through the relationship

$$\lambda = L = ar^*$$

where r^* is the critical radius of the embryo.

a = Bell's parameter

Beginning with the wavelength for the case of a vapor jet surrounded by a liquid, Bell ((5) pg 200) was able to calculate that the "a" parameter for this case would be equal to 6.07 .

Similar calculations show that for the case of a liquid surrounded by a vapor, with $\lambda = (9.02)r_c$, the resulting "a" value is 4.79 .

The actual case encountered in the radiation-induced nucleation studies, is believed to be somewhere between these two cases. That is, the "a" value should lie between 4.79 and 6.07 . (It is probably closer to 6.07, since the jet is more vapor than liquid.)

6.3 Recommendations

Before undertaking any further experimental work, a number of changes are recommended in the experimental apparatus.

The major problem encountered was that of the bubble drifting out of view. The present method of keeping the bubble centered is by means of the convection generator which was discussed in chapter 3. This convection generator, however, was not able to keep the test bubble centered at all times. It proved exceptionally difficult to keep the bubble centered at high temperatures, and many experimental runs were terminated prematurely due to the test bubble drifting out of view.

A new convection generator is recommended. This should have a larger flow-rate capacity and a coolant inlet temperature as low as possible.

Another recommended modification would be a separate recorder for each thermocouple. This would eliminate the switching back and forth on one chart recorder.

It is seriously recommended that a new chamber be constructed. This new chamber should include a larger observation window, preferrably one in which the entire test chamber is visible. It should also include a bright and variable intensity light source, either

inside or outside of the chamber. But it must supply direct illumination of the bubble.

A chamber similar to the one used by T.J. Connolly (31) seems very promising. A sketch of such a type is given in FIG.6.1a . An alternate lighting scheme is shown in FIG.6.1b. Both of these cases must include a convection generator.

The important factors in any new chamber are:

1.) The window to observe the bubble should be as large as possible.

2.) The light source should be as near as possible to the bubble.

3.)A method to keep the bubble in position, such as the convection generator.

Another improvement which can be made, is the degassing of the test liquid. El-Nagdy(4) and Connolly (33) describe a simple method of accomplishing this.

Since it is difficult to find supporting and cover fluids for many test liquids of interest, the concept of a density gradient suspension fluid should be investigated as a possible solution to this problem.

Further experimental work should be done with more organic fluids, preferrably with those for which analytical results have already been obtained.

Different radiation sources and types can be tested, such as gamma rays, or alpha particles.

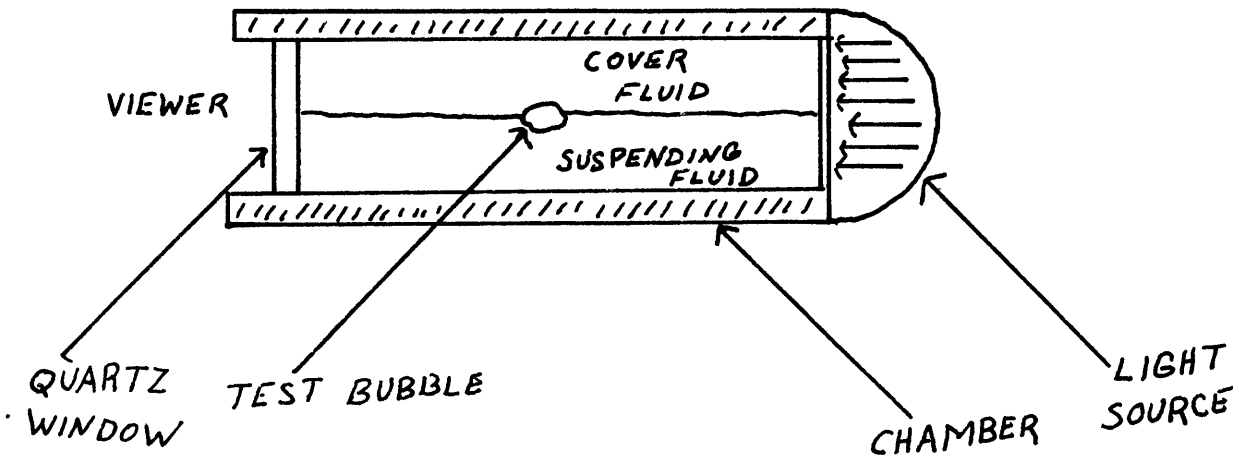


FIG. 6.2a

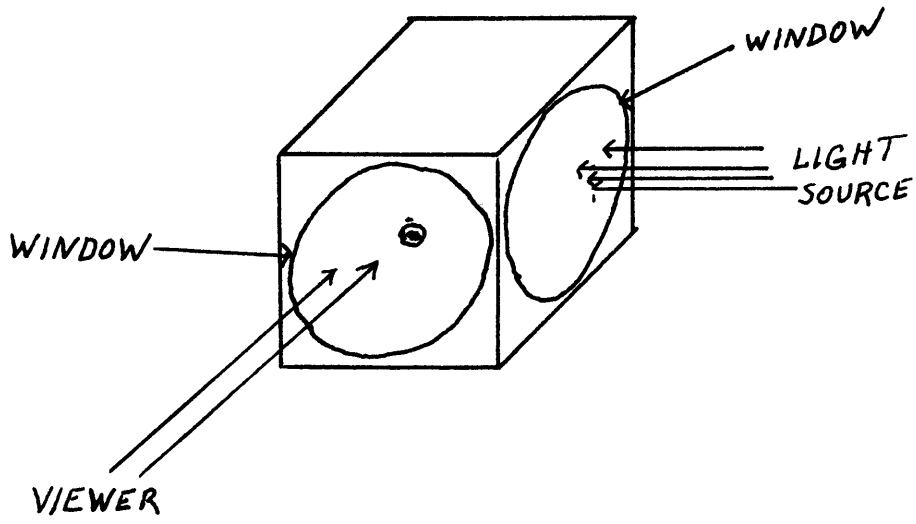


FIG. 6.2b

Theoretical work still remains to be done. It does not seem that the "constant a " theory holds, at least for the cases in which experimental data was obtained. In any case, it has been shown that evenif there is a "constant a " it is not equal to 6.07, but would be somewhat lower.

A possible area of study which might aid in understanding this phenomena, is in the area of the instability and break up of vapor and liquid cylinders, such as the discussion given by Rayleigh.

Appendix A

Physical Properties

A.1 Introduction

Five physical properties of the test liquid are required for the superheat calculations. These are:

1.) Vapor Pressure

2.) Liquid density

3.) Vapor density

4.) Surface tension

5.) Heat of Vaporization

The variation of these properties with temperature must also be known. A search was made to find tabulated data of these properties at various temperatures. This data was then fed into a polynomial regression computer program, which gave an equation for the physical property as a function of temperature. The equations have the following form:

$$P = a + bT + cT^2 + dT^3 + \ldots\ldots\ldots \qquad (A.1)$$

where P = physical property

T = temperature at which the property is evaluated.

a,b,c,d,...are constants determined by the program.

The number of terms required depends on the particular case. The output of the computer program gives the amount of improvement for each additional term added to the polynomial. The first three terms are almost always sufficient.

For some properties a relation between the property and the temperature may be given directly. When this was the case, the equation given was used.

The remainder of this appendix will give the final equations which relate the property to the temperature, and it will also present graphs of the physical property versus the temperature over the temperature range of interest.

Each of the five physical properties have been found for each of the following liquids:

1.) Acetone
2.) Diethyl Ether
3.) Propylene glycol
4.) Nitromethane
5.) Methyl alcohol
6.) Ethyl alcohol

The properties for water and benzene have previously been reported by Bell (5) and Tso (pgs. 84-95 (6)). The properties are listed in the following manner:

- property involved (units) , (reference source)
- temperature dependent equation
- temperature range over which the equation is valid

A.2 Physical Properties of Acetone

1.) Vapor pressure (P_v in mm Hg., T in °C) (22)

$$\log (P_v) = 7.23157 - \frac{1277.03}{T + 237.23}$$

triple point < T < 190 °C

2.) Liquid density (ρ_ℓ lbm/ft^3 T°F) (4)

$$\rho_\ell = 66.3909 - 0.36738(T) + 1.9336 \times 10^{-3} (T)^2 - 3.25478 \times 10^{-6} (T)^3$$

60 °C < T < 230 °C

3.) Vapor density (ρ_v lbm/ft^3 , T°F) (4)

$$\rho_v = 0.45065 - 8.2235 \times 10^{-3}(T) + 3.8308 \times 10^{-5}(T)^2$$

60 °C < T < 230 °C

4.) Surface tension (σ lbf/ft , T°F) (4)

$$\sigma = 1.9457 \times 10^{-3} - 4.8055 \times 10^{-6}(T)$$

60 °C < T < 230 °C

5.) Heat of vaporization (HFG Btu/lbm , T°F) (4)

$$HFG = 251.046 + 0.08274(T) - 0.0017143(T)^2$$

60 °C < T < 230 °C

The graphs of physical property vs temperature for acetone are shown in FIG. A.1 through FIG.A.5 .

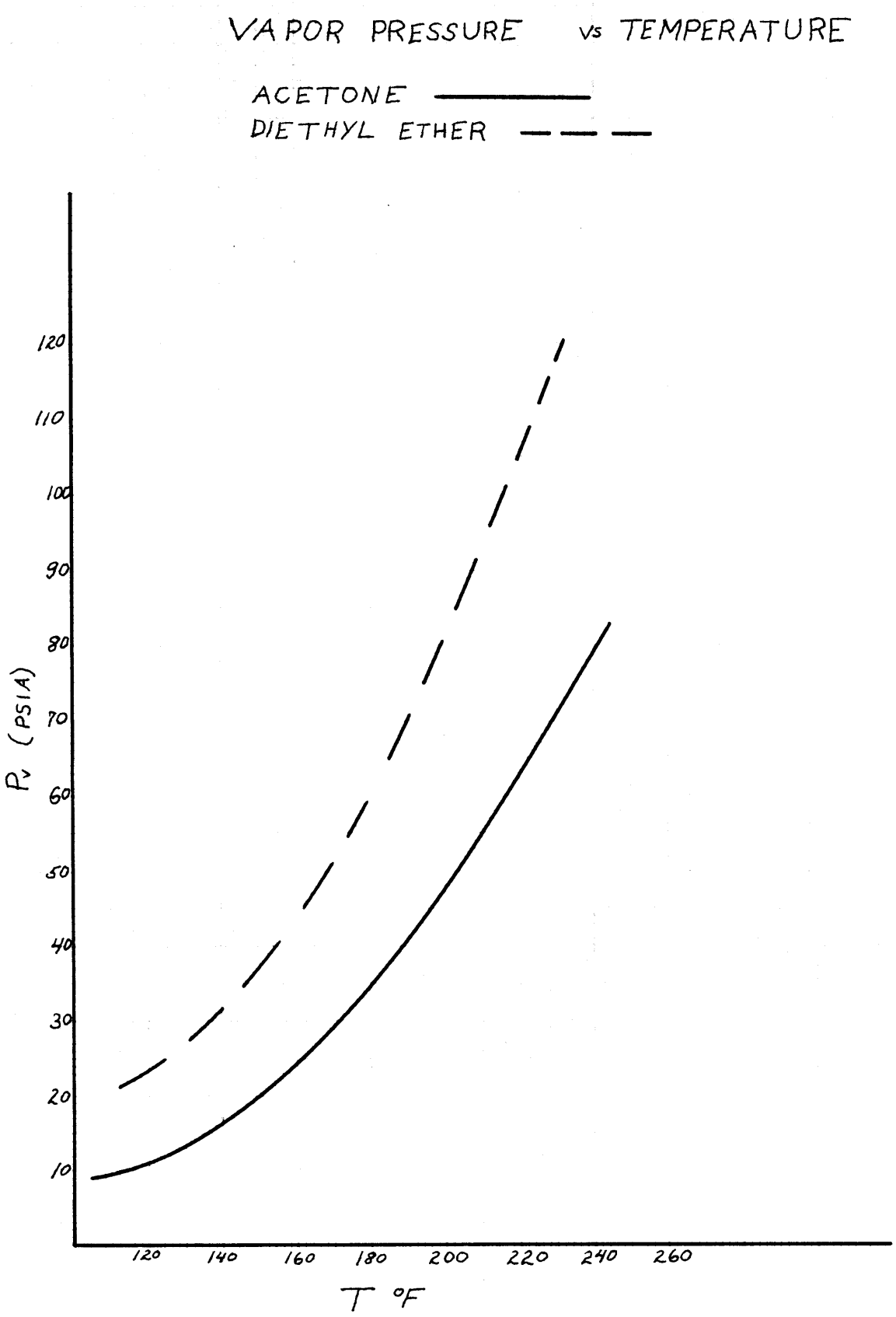

VAPOR PRESSURE vs TEMPERATURE
ACETONE
DIETHYL ETHER
120
110
100
90
80
70
60
50
40
30
20
10
Pv (PSIA)
120
140
160
180
200
220
240
260
T °F

FIG. A.1

VAPOR DENSITY vs TEMPERATURE

ACETONE ——————

DIETHYL ETHER — — —

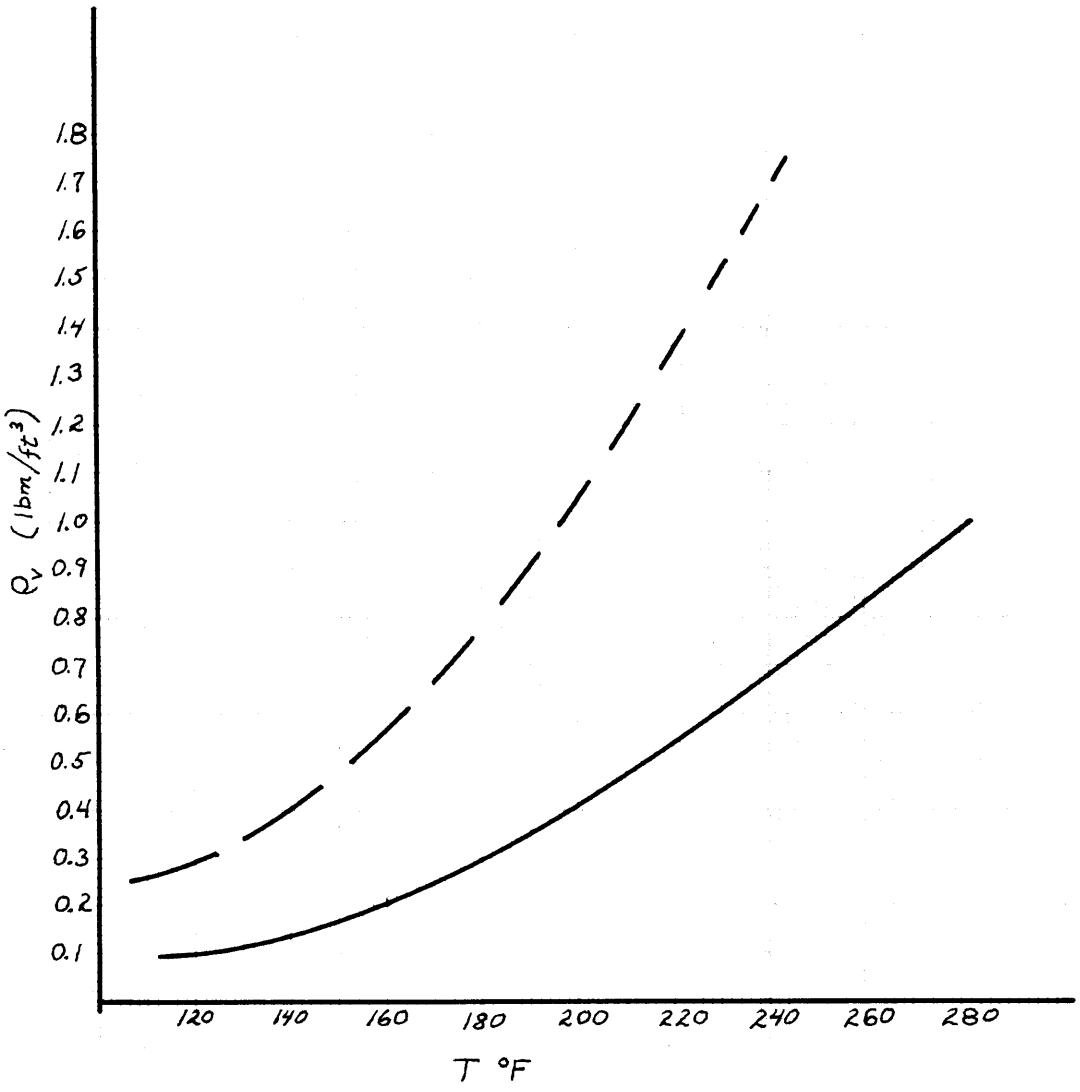


FIG. A.2

LIQUID DENSITY vs TEMPERATURE

ACETONE ——————
DIETHYL ETHER – – – –

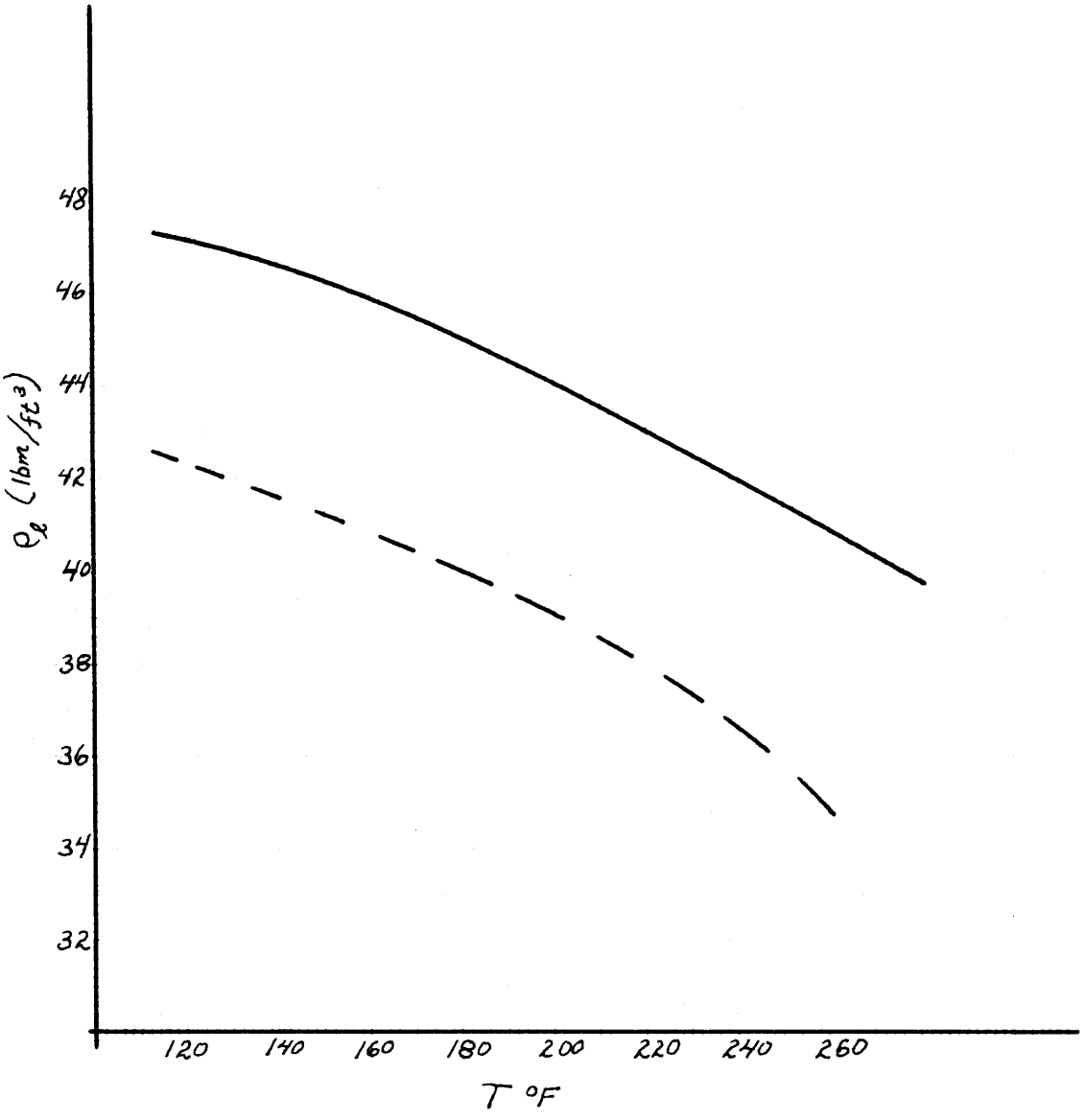


FIG. A.3

SURFACE TENSION vs TEMPERATURE

ACETONE ——————

DIETHYL ETHER — — —

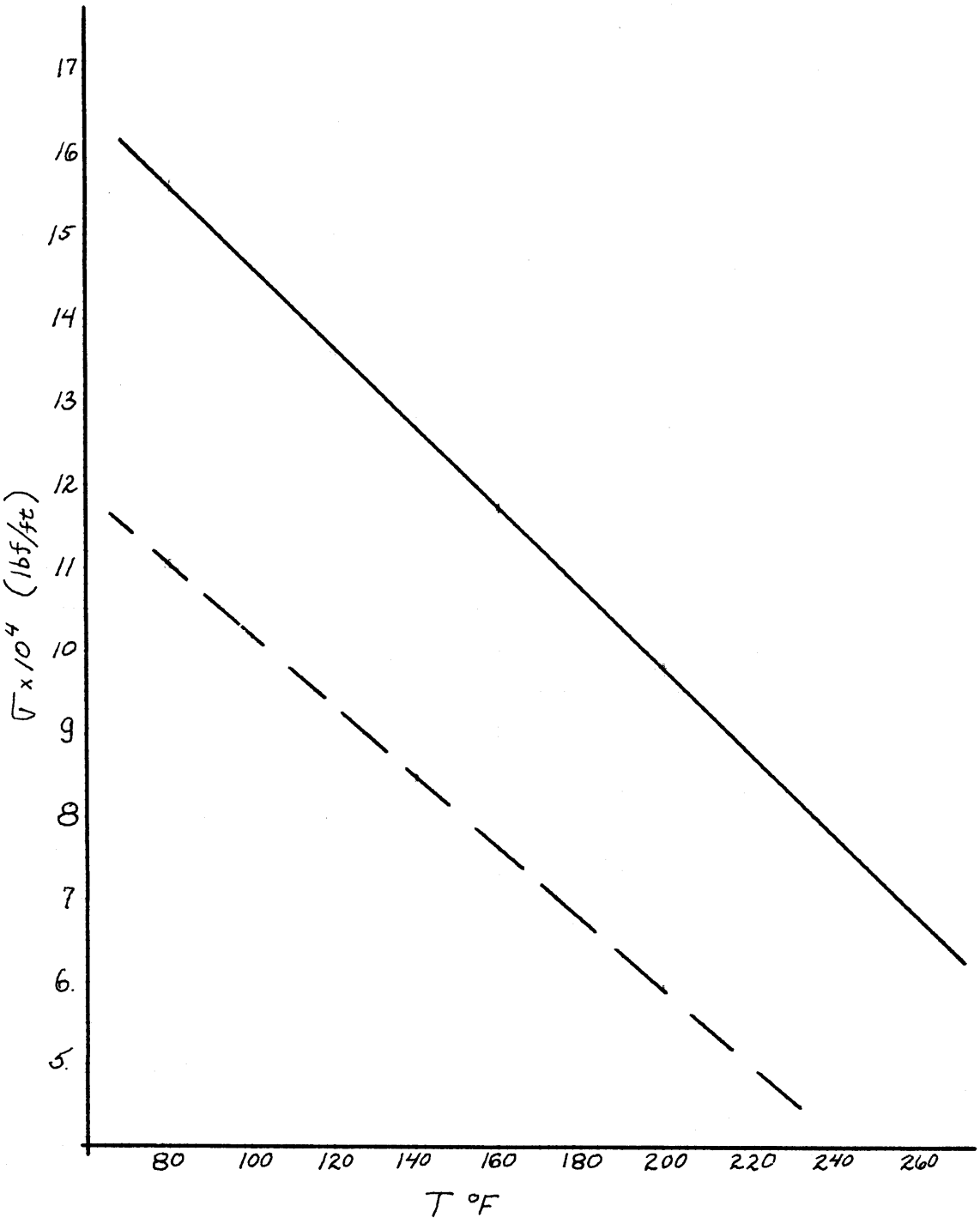


FIG. A.4

HEAT OF VAPORIZATION vs TEMPERATURE

ACETONE ——————
DIETHYL ETHER ——————

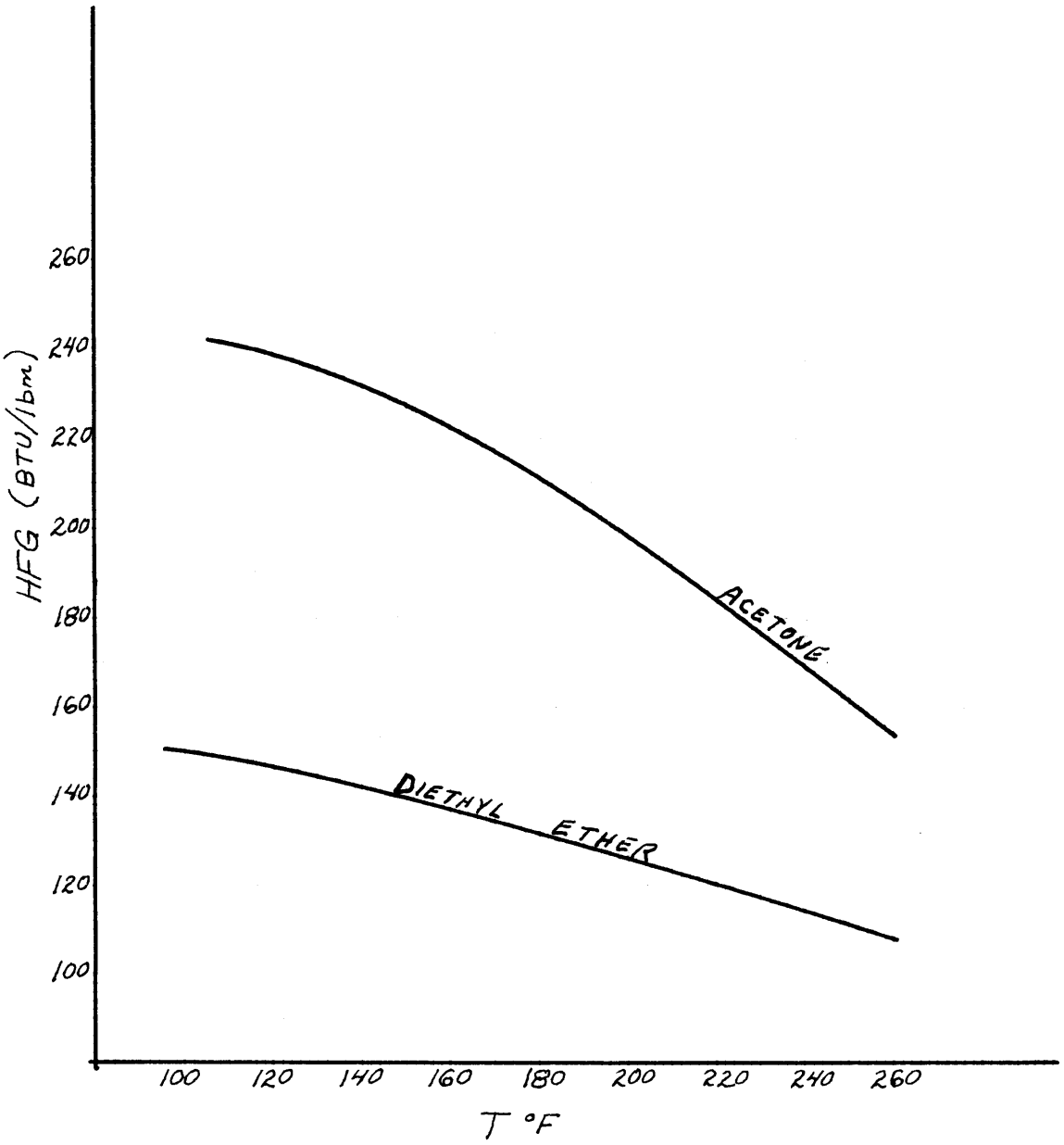


FIG. A.5

A.3 Physical Properties of Diethyl Ether

1.) Vapor pressure (P_v psia , $T^\circ F$) (22)

$$P_v = 44.2626 - 0.96031(T) + 0.0056627(T)^2$$

$$80^\circ F < T < 300^\circ F$$

2.) Liquid density (ρ_ℓ lbm/ft^3 , $T^\circ F$) (29)

$$\rho_\ell = 45.6089 - 0.005339(T) - 0.00013836(T)^2$$

$$65^\circ F < T < 300^\circ F$$

3.) Vapor density (ρ_v lbm/ft^3 , $T^\circ F$) (29)

$$\rho_v = 1.53905 - 0.030649(T) + 0.00013358(T)^2$$

$$65^\circ F < T < 300^\circ F$$

4.) Surface tension (σ lbf/ft , $T^\circ F$)

$$\sigma = 0.0013942 - 3.81359 \times 10^{-6}(T)$$

$$65^\circ F < T < 330^\circ F$$

5.) Heat of vaporization (HFG Btu/lbm , $T^\circ F$) (22)

$$HFG = 162.277 - 0.00444(T) - 8.31636 \times 10^{-4}(T)^2$$

$$80^\circ F < T < 280^\circ F$$

Graphs are shown on FIG. A.1 through A.5 .

A.4 Physical Properties of Propylene Glycol

1.) Vapor pressure (P_v in mm Hg., t°C) (22)

$$\log(P_v) = 8.9171 - \frac{2645.7}{T + 250.7}$$

$$0°C < T < 250°C$$

2.) Liquid density (ρ_ℓ in lbm/ft^3 , T°F) (29)

$$\rho_\ell = 66.3836 - 0.0236(T) - 1.1618\times10^{-5}(T)^2$$

$$20°C < T < 280°C$$

3.) Vapor density (ρ_v in lbm/ft^3 , T°F) (29)

$$\rho_v = 0.2627 - 0.002735(T) + 6.6505\times10^{-6}(T)^2$$

$$20°C < T < 280°C$$

4.) Surface tension (σ in lbf/ft , T°F) (22)

$$\sigma = 0.00285136 - 4.71296\times10^{-6}(T)$$

$$20°C < T < 280°C$$

5.) Heat of vaporization (HFG in Btu/lbm , T°F) (22)

$$HFG = 396.7268 - 0.11884(T) - 3.7196\times10^{-4}(T)^2$$

$$25°C < T < 300°C$$

The graphs of property vs temperature are shown on FIG.A.6 to A.10 .

VAPOR PRESSURE vs TEMPERATURE

PROPYLENE GLYCOL ——————

NITROMETHANE — — — —

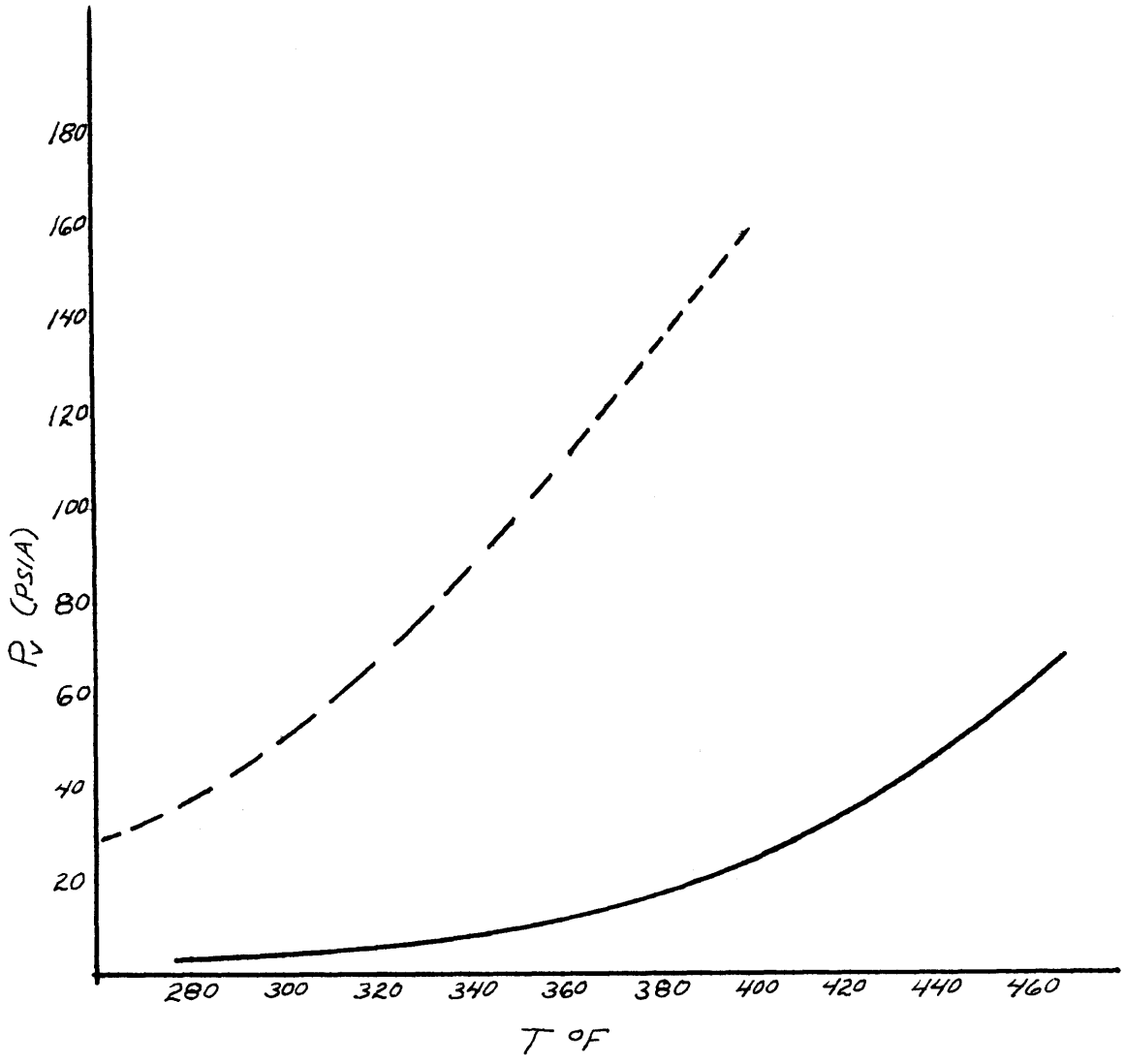


FIG. A.6

LIQUID DENSITY vs TEMPERATURE

PROPYLENE GLYCOL ——————

NITROMETHANE — — — —

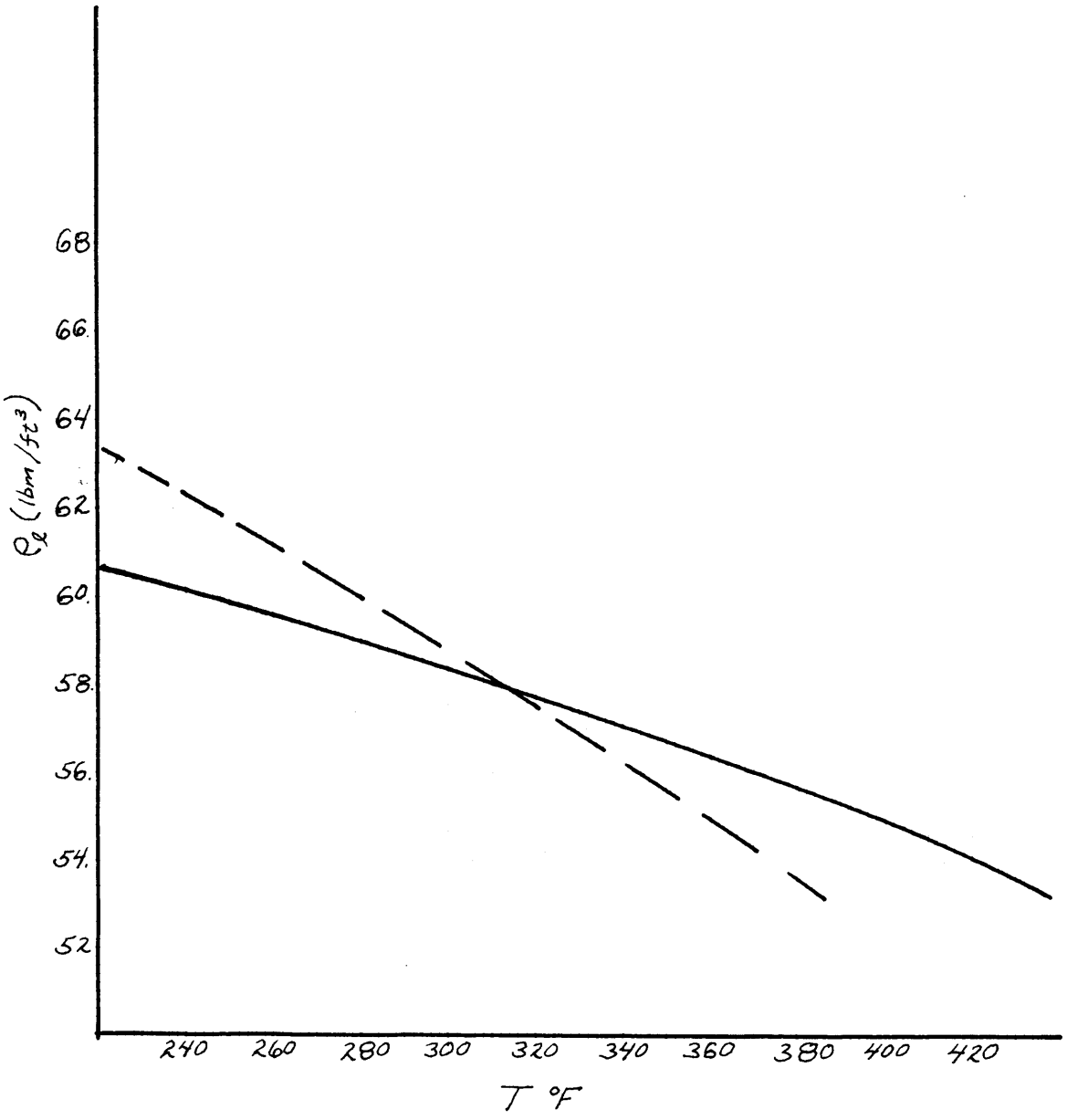


FIG. A.7

VAPOR DENSITY vs TEMPERATURE

PROPYLENE GLYCOL ————————

NITROMETHANE — — — —

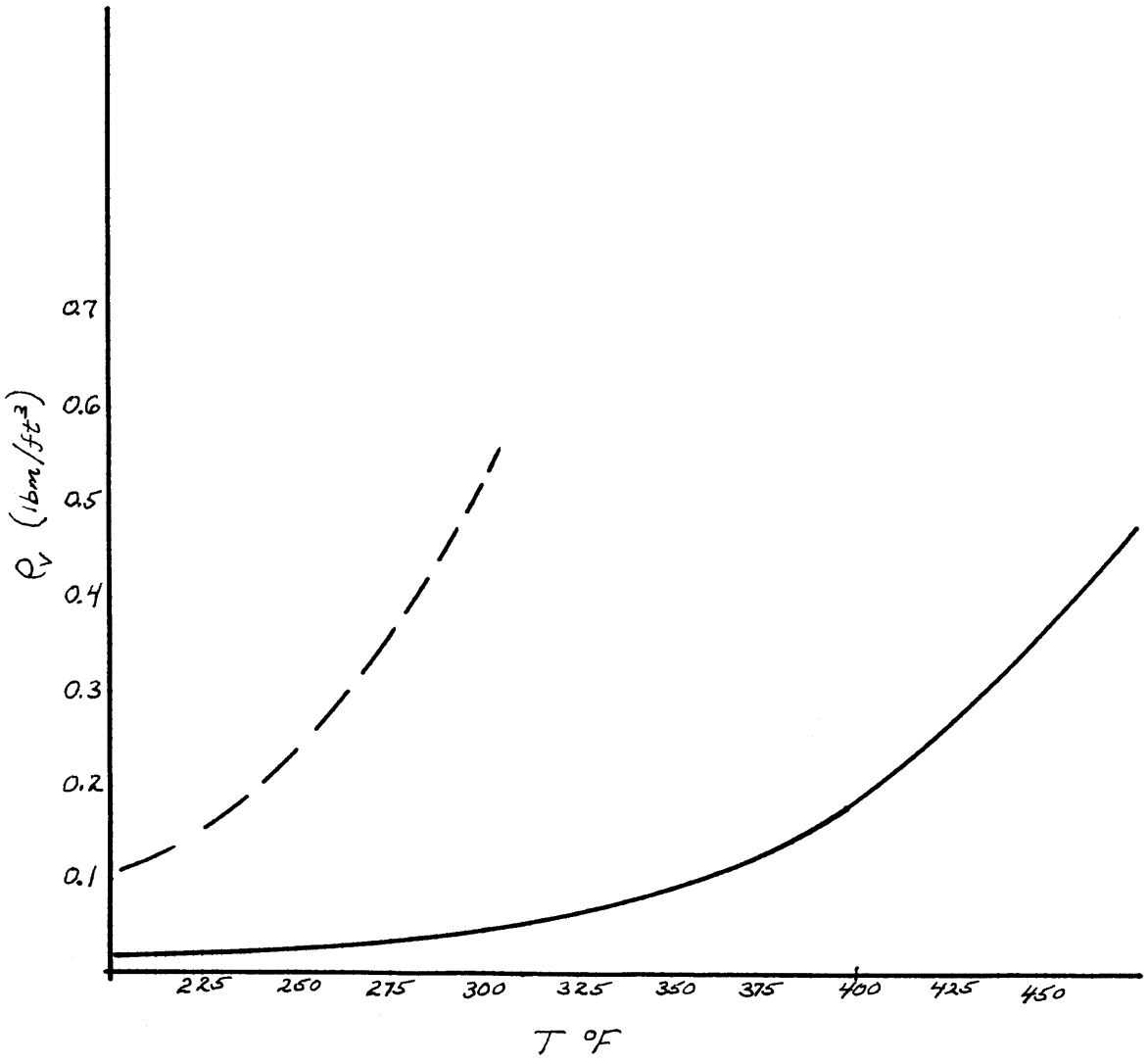


FIG. A.8

SURFACE TENSION vs TEMPERATURE

PROPYLENE GLYCOL ————————

NITROMETHANE — — — —

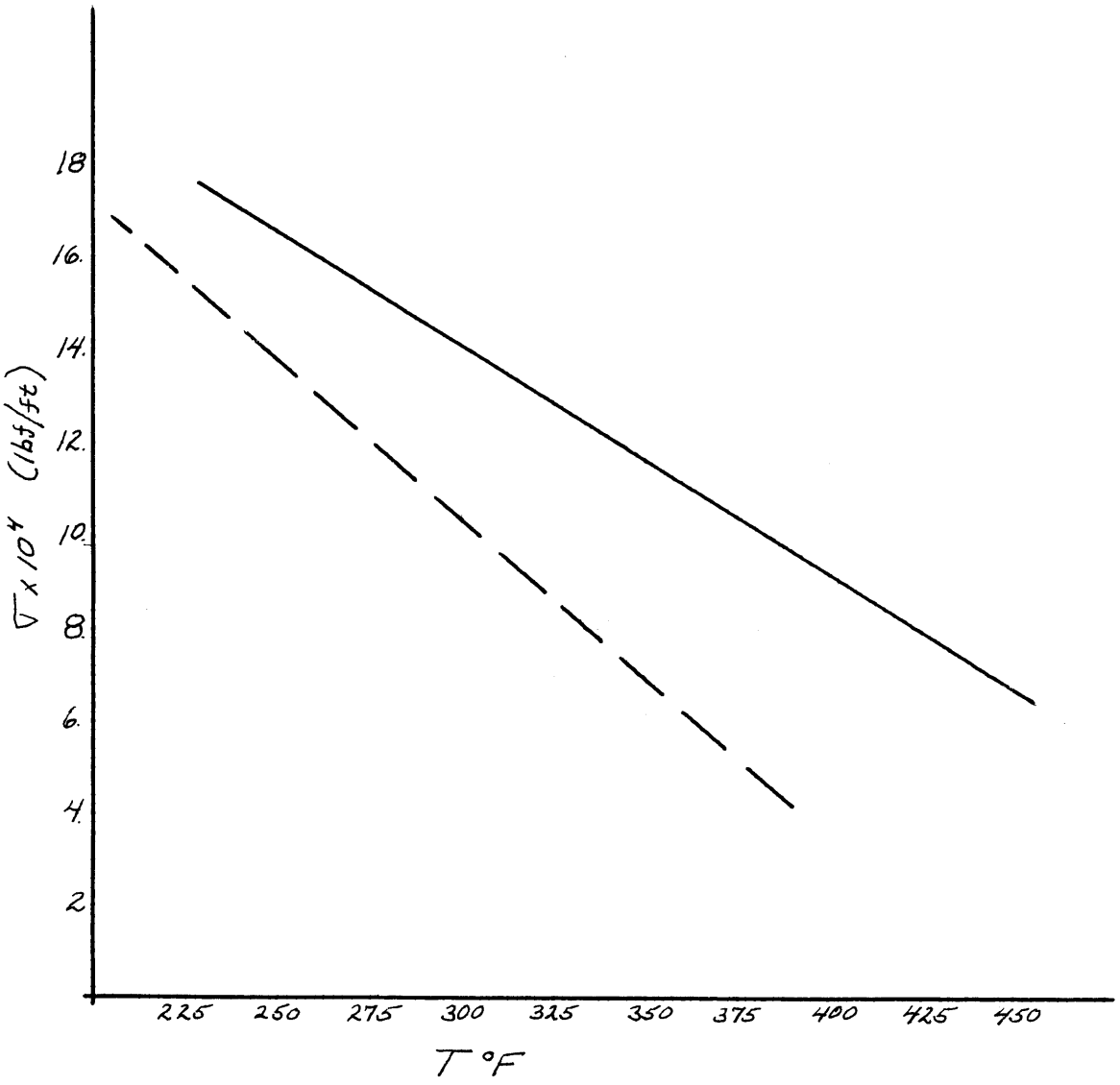


FIG. A.9

HEAT OF VAPORIZATION vs TEMPERATURE

PROPYLENE GLYCOL ——————

NITROMETHANE — — — —

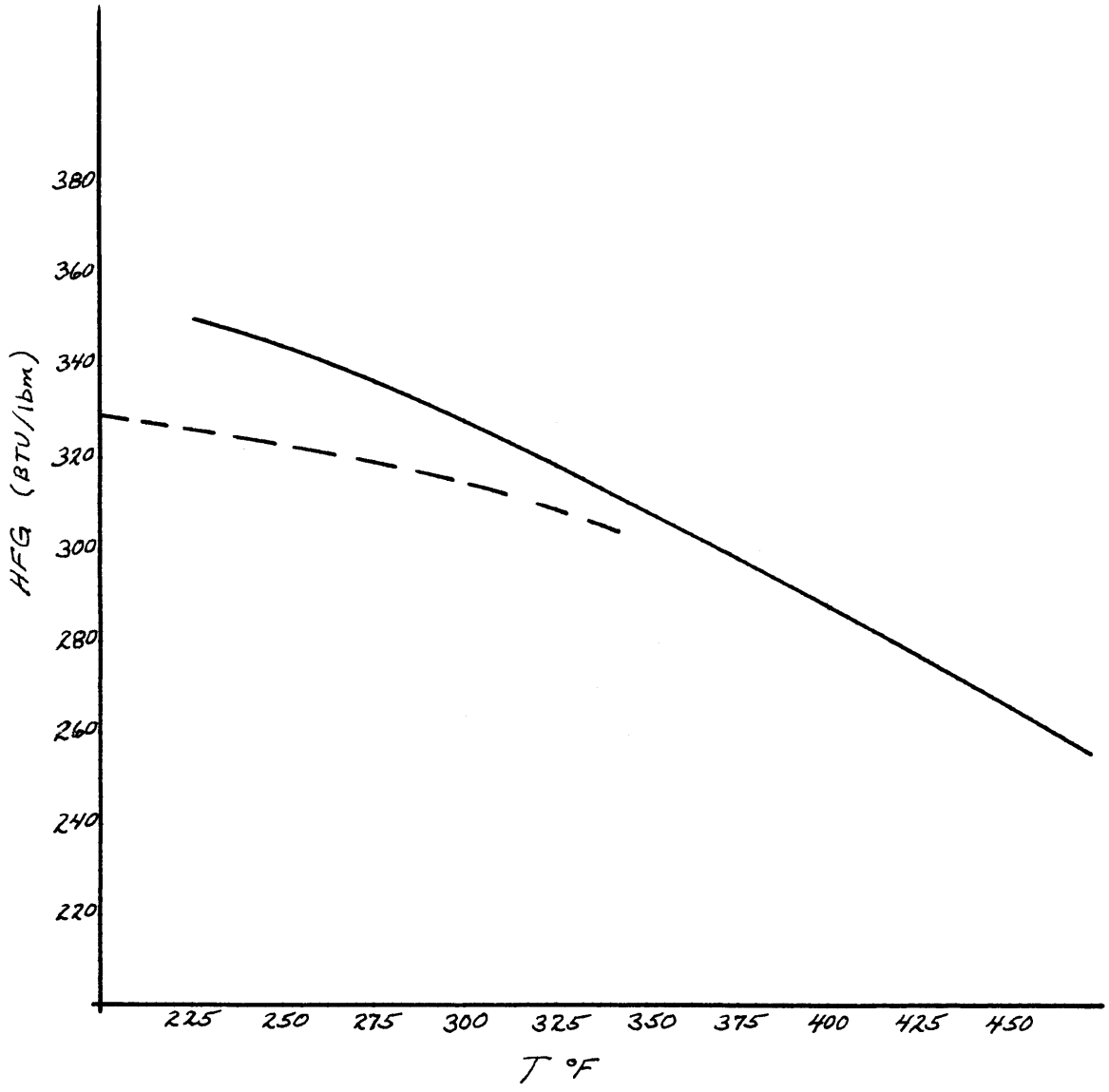


FIG. A.10

A.5 Physical Properties of Nitromethane

1.) Vapor pressure (P_v in psia , T°F) (22)

$$P_v = 19.1127 - 0.342947(T) + 0.0015267(T)^2$$
$$20°C < T < 268°C$$

2.) Liquid density (ρ_ℓ in lbm/ft^3 , T°F) (29)

$$\rho_\ell = 74.07513 - 0.0430263(T) - 2.56308\text{x}10^{-5}(T)^2$$
$$20°C < T < 268°C$$

3.) Vapor density (ρ_v in lbm/ft^3 , T°F) (29)

$$\rho_v = 0.52092 - 0.008079(T) + 2.8548\text{x}10^{-5}(T)^2$$
$$20°C < T < 268°C$$

4.) Surface tension (σ in lbf/ft , T°F) (22)

$$\sigma = 0.0030398 - 6.6158\text{x}10^{-6}(T)$$
$$20°C < T < 268°C$$

5.) Heat of vaporization (HFG in Btu/lbm, T°F) (22)

$$HFG = 343.3718 - 0.009790(T) - 2.5489\text{x}10^{-4}(T)^2$$
$$25°C < T < 265°C$$

The graphs for property vs temperature are found on FIG.A.6 through FIG.A.10 .

A.6 Discussion of Vapor Density Results

Vapor density data for nitromethane was not readily available from the literature. But some investigators have formulated equations which relate the vapor density with the temperature. These are all based on the Verschaffelt equation (30) which has the general form:

$$d_l - d_g = d_o\left(1 - \frac{T}{T_c}\right)^m \qquad (A.2)$$

where; d_l = liquid density

d_g = vapor density

T_c = critical temperature

T = temperature at which the density is to be evaluated.

m, d_o = constants, which differ for different liquids. Density measurements for a number of liquids have shown (29) these equations to be fairly accurate for all temperatures except those close to the critical temperature.

For the case of nitromethane, the general Verschaffelt equation has been broken down (31) into:

$$\log \frac{d}{d_b} = 5.0\left(\frac{T}{T_b} - 1.0\right) \qquad (A.3)$$

where T_b is the liquid boiling point in K , and d_b is the vapor density at the boiling point, which is given by:

$$d_b = \frac{0.0122M}{T_b} \qquad (A.4)$$

where M is the molecular weight.

Another equation for nitromethane, also derived from the basic Verschaffelt equation, is given (29) by:

$$\log d_g = 4.44\left(\frac{T}{T_b} - 1.0\right) + \log \frac{0.0124M}{T_b} \qquad (A.5)$$

where the symbols have the same meaning as in the previous equations.

Calculations were made for the vapor density of nitromethane using both equations (A.3 and A.5). The results obtained from these two equations agreed very well with each other.

These results were then used as input for the polynomial regression program so that a polynomial type equation could be obtained.

This same procedure was used with the propylene glycol vapor density data.

These equations for vapor density may be in error by as much as 5%, (29), but they are the only ones available at this time. Therefore, until more experimental measurements of vapor densities are made, these equations are our best approximations.

A.7 Physical Properties of Methyl Alcohol

1.) Vapor pressure (P_v in psia, T°F) (32)

$$P_v = 98.6548 - 0.018347(T) + 0.00741241(T)^2$$

$$0°C < T < 240°C$$

2.) Liquid density (ρ_ℓ in lbm/ft^3 , T°F) (32)

$$\rho_\ell = 49.5607 - 1.26317\text{x}10^{-4}(T) - 1.02098\text{x}10^{-4}(T)^2$$

$$0°C < T < 240°C$$

3.) Vapor density (ρ_v in lbm/ft^3 , T°F) (29)

$$\rho_v = 0.88826 - 0.0136(T) + 4.7952\text{x}10^{-5}(T)^2$$

$$78°F < T < 375°F$$

4.) Surface tension (σ in lbf/ft , T°F) (32)

$$\sigma = 1.7826\text{x}10^{-3} - 3.4611\text{x}10^{-6}(T)$$

$$0°C < T < 150°C$$

5.) Heat of vaporization (HFG in Btu/lbm, T°F) (32)

$$HFG = 527.091 - 0.1779(T) - 0.0011134(T)^2$$

$$0°C < T < 240°C$$

The graphs for methyl alcohol properties are given in FIG.A.11 through FIG.A.15 .

VAPOR PRESSURE vs TEMPERATURE

METHYL ALCOHOL ———
ETHYL ALCOHOL — — —

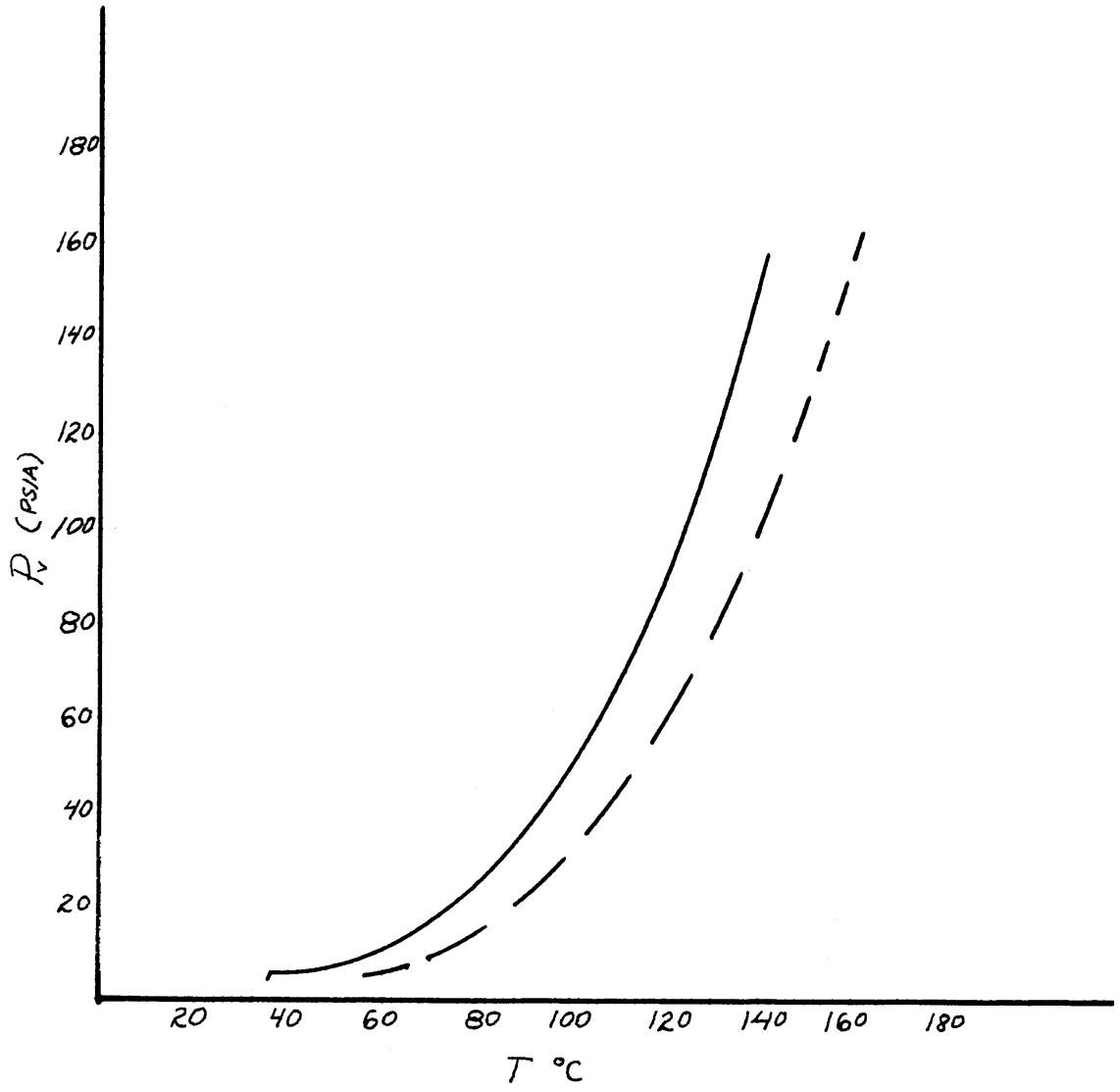


FIG. A. 11

LIQUID DENSITY vs TEMPERATURE

METHYL ALCOHOL ——————

ETHYL ALCOHOL — — — —

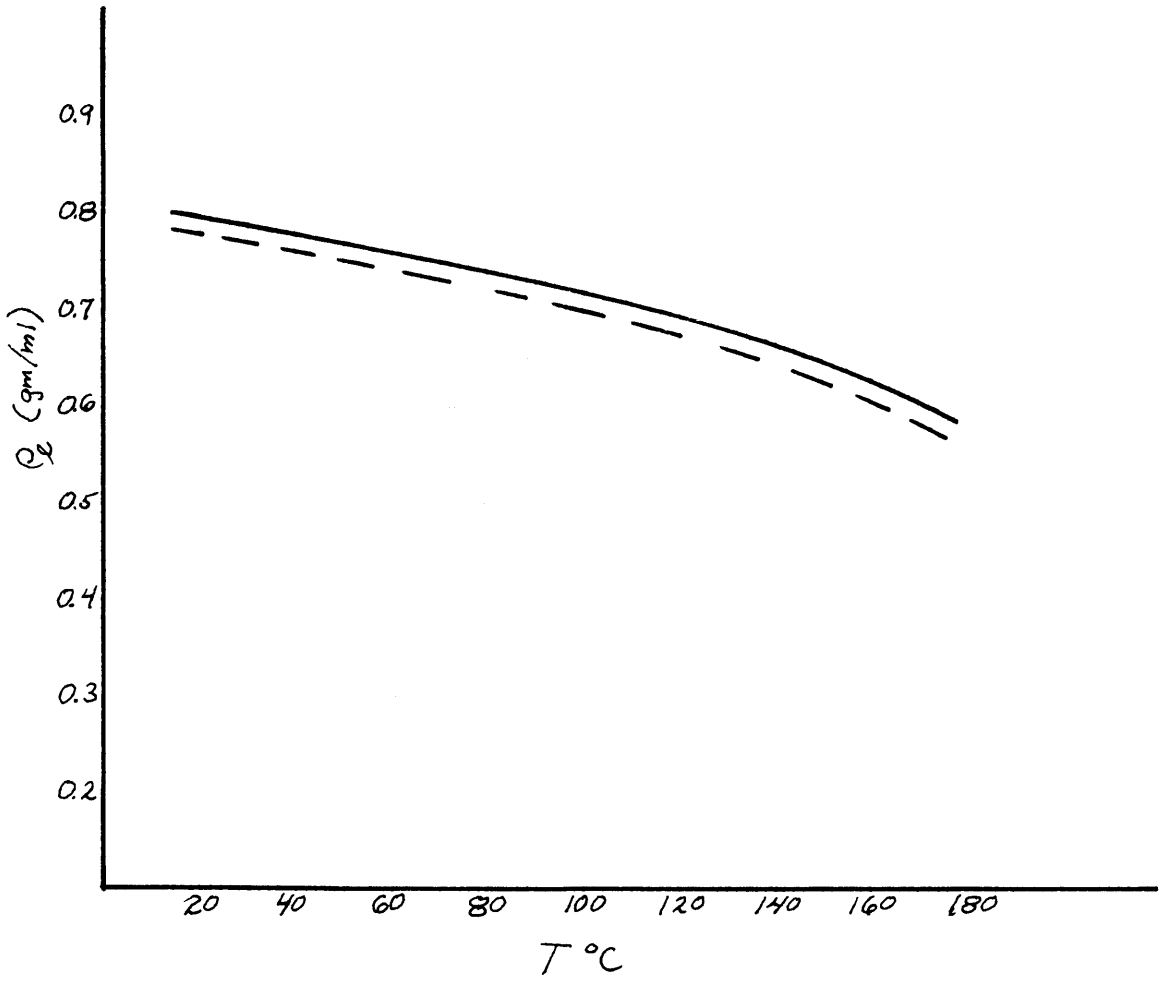


FIG. A. 12

VAPOR DENSITY vs TEMPERATURE

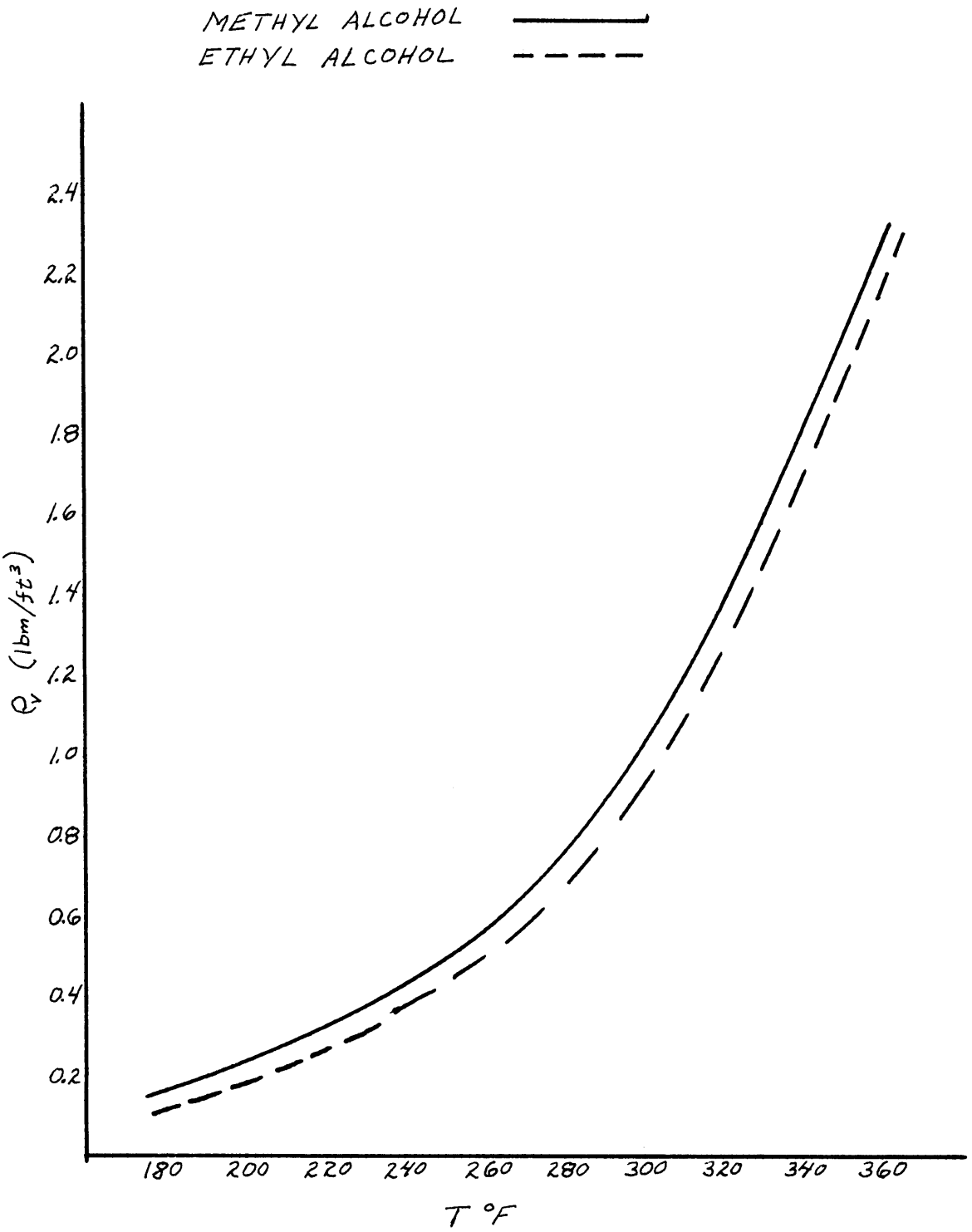


FIG. A.13

SURFACE TENSION vs TEMPERATURE

METHYL ALCOHOL ——————

ETHYL ALCOHOL — — —

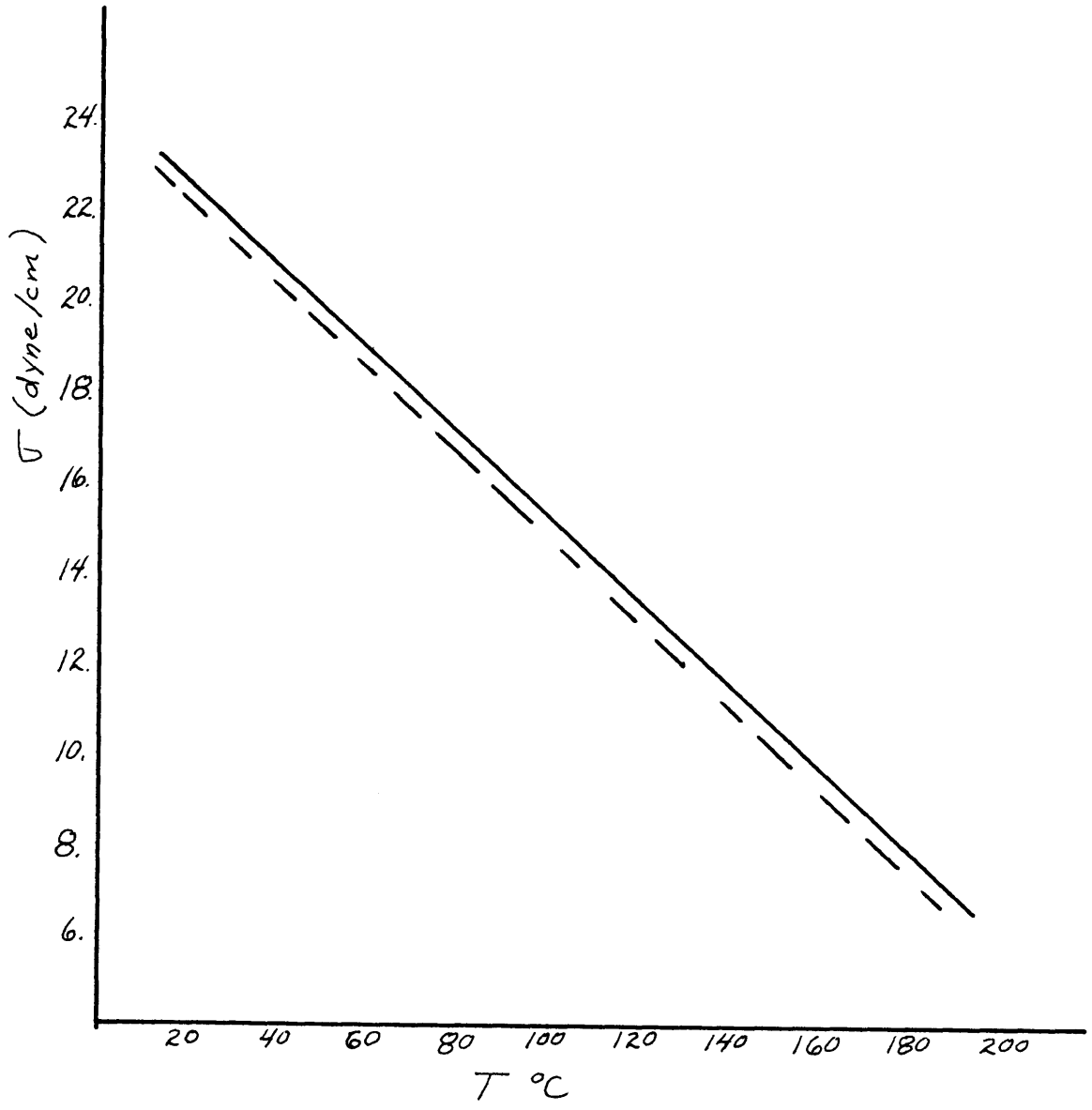


FIG. A. 14

HEAT OF VAPORIZATION vs TEMPERATURE

METHYL ALCOHOL ——————

ETHYL ALCOHOL — — —

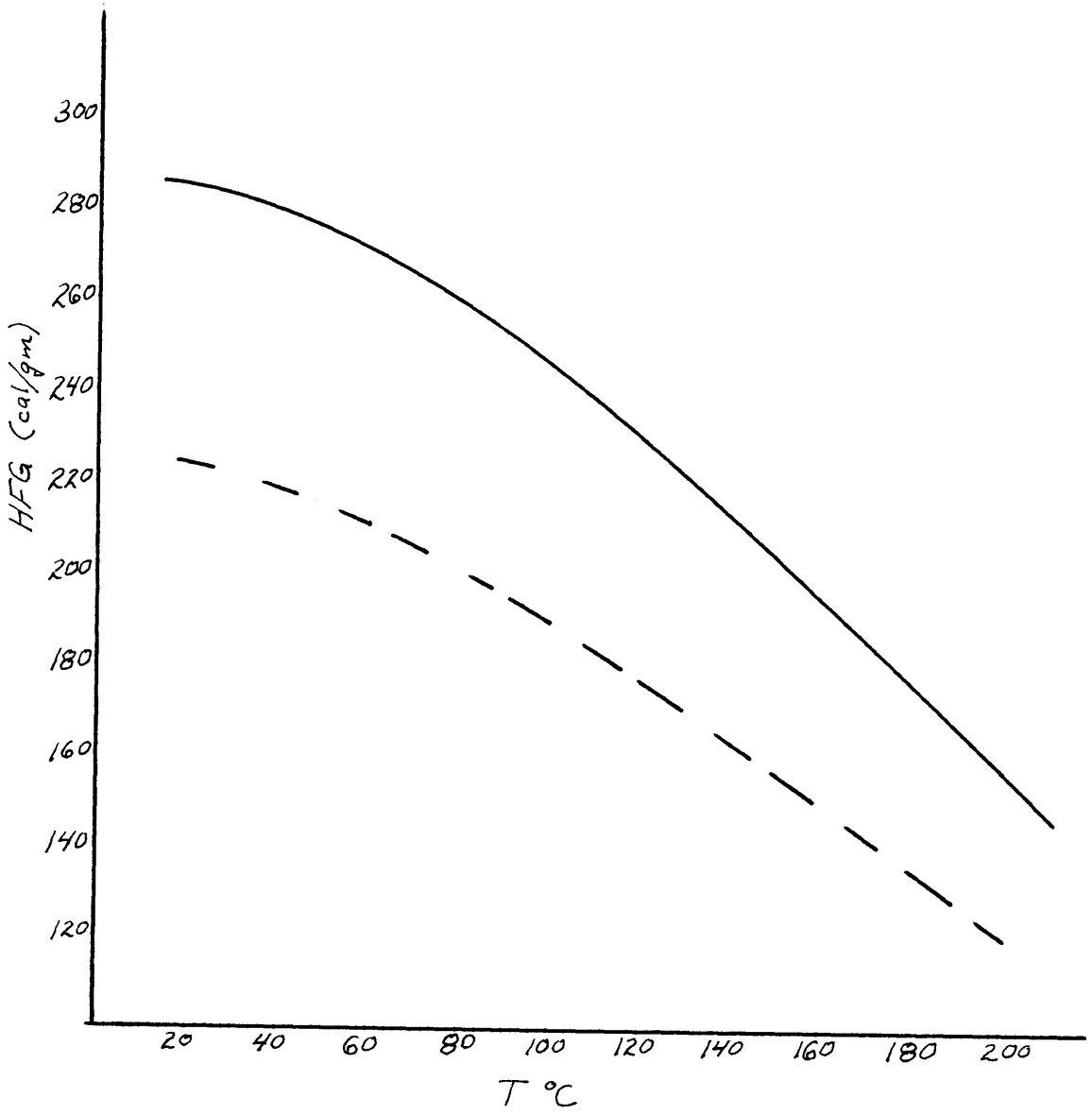


FIG. A. 15

A.8 Physical Properties of Ethyl Alcohol

1.) Vapor pressure (P_v in psia, T°C) (32)

$$P_v = 38.862 - 2.02655(T) + 0.018601(T)^2$$

$$0\,^{\circ}C < T < 240\,^{\circ}C$$

2.) Liquid density (ρ_ℓ in gm/cc , T°C) (32)

$$\rho_\ell = 0.8022 - 5.4322\times10^{-4}(T) - 3.3149\times10^{-6}(T)^2$$

$$0\,^{\circ}C < T < 240\,^{\circ}C$$

3.) Vapor density (ρ_v in gm/cc , T°C) (29)

$$\rho_v = 8.367\times10^{-3} - 3.1762\times10^{-4}(T) + 2.541\times10^{-6}(T)^2$$

$$68\,^{\circ}F < T < 380\,^{\circ}F$$

4.) Surface tension (σ in dynes/cm , T°C) (32)

$$\sigma = 24.5354 - 0.09173(T)$$

$$0\,^{\circ}C < T < 150\,^{\circ}C$$

5.) Heat of Vaporixation (HFG in cal/gm , T°C) (32)

$$HFG = 223.88 - 0.13821(T) - 2.0431\times10^{-3}(T)^2$$

$$0\,^{\circ}C < T < 240\,^{\circ}C$$

The graphs for ethyl alcohol properties are given in FIG.A.11 through FIG.A.15 .

Appendix B

Sample Calculation

Appendix B will give a detailed description of all calculations necessary to calculate the superheat temperature, beginning with the basic equations and concluding with the required inputs for the computer program which performs the actual calculations. The sample calculations will be shown for the case of propylene glycol.

The starting point for all calculations is the energy deposition equation described in chapter 2. This equation is given again below:

$$\frac{1}{N}\frac{dE}{dS} = \frac{4\pi e^4}{m_o V^2}(Z_1)_{eff}^2 \sum_i \nu_i Z_i \ln\left[\frac{1.123\, m_o V^3}{\left(I_i/\hbar\right) e^2 (Z_1)_{eff}}\right]$$
$$+ \frac{4\pi e^4 (Z_1)^2}{V^2} \sum_i \nu_i \frac{Z_i}{M_i} \ln\left[\frac{M_1 M_i V^2 (a_{12}^{scr})_i}{(M_1+M_i) Z_1 Z_i e^2}\right] \qquad \text{(B.1)}$$

All of the terms in equation (B.1) have been described in chapter 2, and will not be repeated here.

The above equation will first be broken down as far as possible without introducing any specific liquid properties.

The second term on the RHS of equation (B.1) can be neglected for all of the cases considered in this work. (The reasons have already been outlined in chapter 2) This leaves only one expression on the RHS. The substitution is now made for $(Z_1)_{eff}$:

$$\frac{1}{N}\frac{dE}{dS} = \frac{4\pi e^4}{m_o v^2}\left[Z_1^{1/3}\frac{\hbar V}{e^2}\right]^2 \sum \nu_i Z_i \ln\left[\frac{1.123\, m_o V^3 \hbar\, e^2}{I_i\, e^2 (Z_1)^{1/3} \hbar V}\right] \quad (B.2)$$

Cancelling all common terms

$$\frac{1}{N}\frac{dE}{dS} = 4\pi (Z_1)^{2/3}\frac{\hbar^2}{m_o}\sum \nu_i Z_i \ln\left[\frac{1.123\, m_o 2E}{I_i (Z_1)^{1/3} M_1}\right] \quad (B.3)$$

where v has been replaced by the energy equivalent, from the basic equation

$$E = \tfrac{1}{2} MV^2 \quad (B.4)$$

The RHS of equation (B.3) is multiplied by $\frac{c^2}{c^2}$,(or 1) resulting in

$$\frac{1}{N}\frac{dE}{dS} = 4\pi (Z_1)^{2/3}\frac{(\hbar c)^2}{m_o c^2}\sum \nu_i Z_i \ln\left[\frac{1.123\, m_o 2E}{I_i (Z_1)^{1/3} M_1}\right] \quad (B.5)$$

This form gives the constants (such as h,c, m_o) in familiar forms, the values of which can be found in any nuclear physics text. More specifically, we have:

$hc = 1.9732 \times 10^{-11}$ Mev-cm

$m_o c^2 = 0.511$ Mev.

$m_o = 0.5488 \times 10^{-3}$ amu.

The N term is replaced by

$$N = \frac{\rho_\ell N_{av}}{M_{MEDIUM}} \qquad (B.6)$$

and brought to the RHS giving

$$\frac{dE}{dS} = \frac{\rho_\ell N_{av}}{M_{MEDIUM}} 4\pi (Z_1)^{2/3} \frac{(1.9732 \times 10^{-11})^2}{0.511} \sum_i \nu_i Z_i \ln \left[\frac{1.232 \times 10^{-3} E}{I_i (Z_1)^{1/3} M_1} \right] \qquad (B.7)$$

This is as far as the original equation, (B.1), can be broken down without introducing any specific particle or medium. The case of fission fragments in propylene glycol will now be taken as an example.

The fission fragment properties are as follows ((5) pg 115):

$$A = 97 = M_1$$

$$Z = 38 = Z_1$$

The molecular weight of the medium is

$$M_{medium} = M_{\substack{propylene \\ glycol}} = 76.096$$

Substituting the values for M_1, Z_1, M_{medium}, and for $N_{av} = 6.023 \times 10^{-23}$, equation (B.7) becomes:

$$\frac{dE}{dS} = (850.0) \rho_\ell \sum_i \nu_i Z_i \ln \left[\frac{(3.777 \times 10^{-6}) E}{I_i} \right] \qquad (B.8)$$

The summation term is over the atoms in the stopping medium (propylene glycol in this case). The chemical formula for propylene glycol is $C_3H_8O_2$, so the summation sign can be shown as follows: $\sum_{i=C,H,O}$ The values for ν_i, Z_i , and I_i are:

$\nu_C = 3$	$Z_C = 6$	$I_C = 76.4$ev
$\nu_H = 8$	$Z_H = 1$	$I_H = 15.5$ ev
$\nu_O = 2$	$Z_O = 8$	$I_O = 100.0$ ev

Values for the mean ionization potential, I_i , have been obtained from Evans (14).

The values for I_i are given in ev, but the I_i term in equation (B.8) must be in units of Mev (E is given in Mev).

Equation (B.8) thus becomes:

$$\frac{dE}{ds} = \rho_e(850.0)\left[(3)(6)\ln\left(\frac{3.777\times10^{-6}E}{76.4\times10^{-6}}\right) + (8)(1)\ln\left(\frac{3.777\times10^{-6}E}{15.5\times10^{-6}}\right) + (2)(8)\ln\left(\frac{3.777\times10^{-6}E}{100.0\times10^{-6}}\right)\right] \quad \text{(B.9)}$$

Equation (B.9) is now reduced to an equation with a single "ln" term:

$$\frac{dE}{dS} = \rho_\ell \left(3.57 \times 10^4\right) \ln\left(0.0604 E\right) \qquad \text{(B.10)}$$

Equation (B.10) is the final form of the energy deposition equation.

dE/dS is in terms of Mev/cm

ρ_ℓ in terms of gm/cc

E in terms of Mev.

The remainder of the calculations provide input parameters to the computer program.

Equation (B.10) is substituted into the integral equation described in chapter 2,

$$\frac{-\left[E(S_1) - E(S_2)\right]^2}{a r^*} = \int_{E(S_1)}^{E(S_2)} \left(\frac{dE}{dS}\right) dE \qquad \text{(B.11)}$$

Performing the integration:

$$\frac{-\left[E(S_1) - E(S_2)\right]^2}{a r^*} = \frac{\rho_\ell \left(3.57 \times 10^4\right)}{0.0604} \left[0.0604 E \ln 0.0604 E - 0.0604 E\right]_{E(S_1)}^{E(S_2)}$$

(B.12)

Further breakdown of (B.12) and combination of terms leads to:

$$\frac{\left[E(s_1) - E(s_2)\right]^2}{E(s_1)\ln E(s_1) - E(s_2)\ln E(s_2) - 3.81\left(E(s_1) - E(s_2)\right)} = a r^* \rho_\ell \left(3.57 \times 10^4\right) \qquad (B.13)$$

The RHS of equation (B.13) is in metric units and is now converted to allow ρ_ℓ and r^* to be input in English units. The final conversion factor is 2.0479, and the RHS must be divided by this. (The LHS is in Mev terms and must remain the same numerical value, no matter what units the RHS has.)

Converting to English units and substituting the relationship,

$$r^* = \frac{2\sigma}{P_v + P_c - P_\ell} \qquad (B.14)$$

the RHS of equation (B.13) becomes:

$$\frac{\left(3.486 \times 10^4\right) a \sigma \rho_\ell}{P_v + P_c - P_\ell} \qquad (B.15)$$

The expression in (B.15) is one of the parameters which is used as an input to the computer program. This parameter is given the name of "COEF".

$$\text{COEF} \equiv \frac{\left(3.486 \times 10^4\right) a \sigma \rho_\ell}{P_v + P_c - P_\ell} \qquad (B.16)$$

COEF is now the RHS of equation (B.13) , which is written as

$$\frac{\left[E(S_1)-E(S_2)\right]^2}{E(S_1)\ln E(S_1) - E(S_2)\ln E(S_2) - 3.81\left(E(S_1)-E(S_2)\right)} = COEF \qquad (B.17)$$

Rearranging terms in equation (B.17)

$$E(S_1)^2 - COEF\cdot E(S_1)\left[\ln E(S_1) - 3.81\right] = E(S_2)\left[2\cdot E(S_1) + 3.81\cdot COEF\right] - E(S_2)\cdot COEF - \ln E(S_2) - E(S_2)^2 \qquad (B.18)$$

The term enclosed in brackets on the RHS is another input to the program, and is labeled "DDD" .

$$DDD \equiv \left[2E(S_1) + 3.81\cdot COEF\right] \qquad (B.19)$$

The LHS of equation (B.18) can be rewritten in a slightly different form, and is also used as an input to the program. The LHS is labeled "CCC" .

$$CCC \equiv E(S_1)^2 - COEF\cdot E(S_1)\left[\ln 0.0604\,E(S_1) - 1.0\right] \qquad (B.20)$$

Equation (B.18) can thus be written as

$$CCC = E(S_2)\left[DDD - COEF \cdot \ln E(S_2)\right] - E(S_2)^2 \qquad (B.21)$$

The term inside of the brackets is also an input, and is labeled "BBB" .

$$BBB \equiv DDD - COEF \cdot \ln E(S_2) \qquad (B.22)$$

There are thus four inputs to the computer program which were derived from the original energy deposition formula. These were:

1.) COEF

2.) CCC

3.) DDD

4.) BBB

These terms will be different for different fluids.

For the case of fast neutrons in the medium, the only difference is that the subscript 1 in equation (B.2) refers to the PKOA. In this case there are actually three possible PKOAs, namely H , C, and O. But, as has been shown by Bell (5), and by El-Nagdy(4)(who went through minimum superheat calculations for each type of atom), the minimum superheat will be obtained with the O-PKOA. Therefore, only the O (Oxygen) PKOA is considered.

There are some more computer inputs which should be discussed. (The computer programs are listed in Appendix C.). One input is the term labeled "EPKA". This is the initial energy of the radiation particle, which has been carried in the equations as $E(s_1)$. The value for $E(s_1)$ will always be known.

The physical properties as a function of temperature are also inputs. These have been given in Appendix A .

There are two final inputs which must be considered. These are computer statement number

```
0558 (IF(ABS(E-EC)-X.XXX)9858,0658,0658
```

and the statement

```
E = Y.YYYY
```

The important point here is the value of X.XXX in statement 0558. This value sets the convergence criteria. If this convergence criteria is too strict, a COMPLETION CODE error will result. If this is the case, then the convergence criteria must be relaxed (X.XXX must be made larger).

The best method of determining what the value of X.XXX should be, is to choose values for E (beginning with $E(s_1)$ and gradually decreasing in energy) and actually calculating the value of EC.

EC is calculated in statement number 0458:

```
0458 EC = E -(E*E-BBB*E+CCC)/(2.0*E-BBB)
```

A simple computer program can be written to calculate EC values for various E values. This will then give an indication of the minimum value for the ABS(E-EC) term used in statement 0558. This minimum value is then used as the value for X.XXX . (For the present work the value of X.XXX varied from 0.0001 to 0.003)

The value chosen for the E term in

$$E = Y.YYY$$

is not too critical, and an approximate value which can be used is : $E = \frac{1}{2}E(s_1)$.

These (from COEF to E) are the factors in the computer program which must be changed for the different fluids. Otherwise, the same basic program may be used for all cases.

Appendix C

Computer Programs

This Appendix contains the three basic computer programs used in this work.

The first one given is the Polynomial Regression Program which gives us the equations for the physical properties as a function of temperature. (Nitromethane is shown as an example.)

The seoond program listed in this Appendix is the program used to calculate the superheat threshold for the case of fission fragments. (Benzene parameters are used as an example.)

The third program is for calculating the superheat threshold for fast neutrons. (Benzene parameters are used as an example.)

```
C                                                                       POLY0001
C                                                                       POLY0002
C     ***************************************************************** POLY0003
C     *                                                               * POLY0004
C     *                                                               * POLY0005
C     *                                                               * POLY0006
C     *              POLYNOMIAL REGRESSION PROGRAM                    * POLY0007
C     *                                                               * POLY0008
C     *                                                               * POLY0009
C     *                                                               * POLY0010
C     ***************************************************************** POLY0011
C                                                                       POLY0012
C                                                                       POLY0013
C      NITROMETHANE PROPERTIES   --  ENGLISH UNITS  -----               POLY0014
C                                                                       POLY0015
C             SURFACE TENSION        LBF/FT      @  DEG F              POLY0016
C                                                                       POLY0017
C             VAPOR PRESSURE       PSIA          @   DEG F             POLY0018
C                                                                       POLY0019
C             HEAT OF VAPORIZATION         BTU/LBM      @  DEG F       POLY0020
C                                                                       POLY0021
C             LIQUID DENSITY        LBM/FT3      @  DEG F              POLY0022
C                                                                       POLY0023
C             VAPOR DENSITY           LBM/FT3       @  DEG F          POLY0024
C                                                                       POLY0025
C                                                                       POLY0026
C                                                                       POLY0027
C     ...............................................................   POLY0028
C                                                                       POLY0029
C        SAMPLE MAIN PROGRAM FOR POLYNOMIAL REGRESSION - POLRG          POLY0030
C                                                                       POLY0031
C        PURPOSE                                                        POLY0032
C           (1) READ THE PROBLEM PARAMETER CARD FOR A POLYNOMIAL REGRES- POLY0033
C           SION, (2) CALL SUBROUTINES TO PERFORM THE ANALYSIS, (3)     POLY0034
C           PRINT THE REGRESSION COEFFICIENTS AND ANALYSIS OF VARIANCE  POLY0035
C           TABLE FOR POLYNOMIALS OF SUCCESSIVELY INCREASING DEGREES,   POLY0036
```

```
C          AND (4) OPTIONALLY PRINT THE TABLE OF RESIDUALS AND A PLOT      POLY0037
C          OF Y VALUES AND Y ESTIMATES.                                    POLY0038
C       REMARKS                                                            POLY0039
C          THE NUMBER OF OBSERVATIONS, N, MUST BE GREATER THAN M+1,        POLY0040
C          WHERE M IS THE HIGHEST DEGREE POLYNOMIAL SPECIFIED.             POLY0041
C          IF THERE IS NO REDUCTION IN THE RESIDUAL SUM OF SQUARES         POLY0042
C          BETWEEN TWO SUCCESSIVE DEGREES OF THE POLYNOMIALS, THE          POLY0043
C          PROGRAM TERMINATES THE PROBLEM BEFORE COMPLETING THE ANALY-     POLY0044
C          SIS FOR THE HIGHEST DEGREE POLYNOMIAL SPECIFIED.                POLY0045
C                                                                          POLY0046
C       SUBROUTINES AND FUNCTION SUBPROGRAMS REQUIRED                      POLY0047
C          GDATA                                                           POLY0048
C          ORDER                                                           POLY0049
C          MINV                                                            POLY0050
C          MULTR                                                           POLY0051
C          PLOT   (A SPECIAL PLOT SUBROUTINE PROVIDED FOR THE SAMPLE       POLY0052
C                 PROGRAM.)                                                POLY0053
C                                                                          POLY0054
C       METHOD                                                             POLY0055
C          REFER TO B. OSTLE, 'STATISTICS IN RESEARCH', THE IOWA STATE     POLY0056
C          COLLEGE PRESS', 1954, CHAPTER 6.                                POLY0057
C                                                                          POLY0058
C     ..............................................................       POLY0059
C                                                                          POLY0060
C     THE FOLLOWING DIMENSION MUST BE GREATER THAN OR EQUAL TO THE         POLY0061
C     PRODUCT OF N*(M+1), WHERE N IS THE NUMBER OF OBSERVATIONS AND M      POLY0062
C     IS THE HIGHEST DEGREE POLYNOMIAL SPECIFIED..                         POLY0063
C                                                                          POLY0064
      DIMENSION X(1100)                                                    POLY0065
C                                                                          POLY0066
C     THE FOLLOWING DIMENSION MUST BE GREATER THAN OR EQUAL TO THE         POLY0067
C     PRODUCT OF M*M..                                                     POLY0068
C                                                                          POLY0069
      DIMENSION DI(100)                                                    POLY0070
C                                                                          POLY0071
C     THE FOLLOWING DIMENSION MUST BE GREATER THAN OR EQUAL TO             POLY0072
```

```
C      (M+2)*(M+1)/2..                                                  POLY0073
C                                                                       POLY0074
         DIMENSION D(66)                                                POLY0075
C                                                                       POLY0076
C      THE FOLLOWING DIMENSIONS MUST BE GREATER THAN OR EQUAL TO M..    POLY0077
C                                                                       POLY0078
         DIMENSION B(10),E(10),SB(10),T(10)                             POLY0079
C                                                                       POLY0080
C      THE FOLLOWING DIMENSIONS MUST BE GREATER THAN OR EQUAL TO (M+1).. POLY0081
C                                                                       POLY0082
         DIMENSION XBAR(11),STD(11),COE(11),SUMSQ(11),ISAVE(11)         POLY0083
C                                                                       POLY0084
C      THE FOLLOWING DIMENSION MUST BE GREATER THAN OR EQUAL TO 10..    POLY0085
C                                                                       POLY0086
         DIMENSION ANS(10)                                              POLY0087
C                                                                       POLY0088
C      THE FOLLOWING DIMENSION WILL BE USED IF THE PLOT OF OBSERVED DATA POLY0089
C      AND ESTIMATES IS DESIRED.  THE SIZE OF THE DIMENSION, IN THIS    POLY0090
C      CASE, MUST BE GREATER THAN OR EQUAL TO N*3.  OTHERWISE, THE SIZE POLY0091
C      OF DIMENSION MAY BE SET TO 1.                                    POLY0092
C                                                                       POLY0093
         DIMENSION P(300)                                               POLY0094
C                                                                       POLY0095
C      .................................................................POLY0096
C                                                                       POLY0097
C         IF A DOUBLE PRECISION VERSION OF THIS ROUTINE IS DESIRED, THE POLY0098
C         C IN COLUMN 1 SHOULD BE REMOVED FROM THE DOUBLE PRECISION     POLY0099
C         STATEMENT WHICH FOLLOWS.                                      POLY0100
C                                                                       POLY0101
C      DOUBLE PRECISION X,XBAR,STD,D,SUMSQ,DI,E,B,SB,T,ANS,DET,COE      POLY0102
C                                                                       POLY0103
C         THE C MUST ALSO BE REMOVED FROM DOUBLE PRECISION STATEMENTS   POLY0104
C         APPEARING IN OTHER ROUTINES USED IN CONJUNCTION WITH THIS     POLY0105
C         ROUTINE.                                                      POLY0106
C                                                                       POLY0107
C         .............................................................POLY0108
```

```
C
    1 FORMAT(A4,A2,I5,I2,I1)                                              POLY0109
    2 FORMAT(2F10.0)                                                      POLY0110
    3 FORMAT(27H1POLYNOMIAL REGRESSION.....,A4,A2/)                       POLY0111
    4 FORMAT(23HONUMBER OF OBSERVATIONS,I6//)                             POLY0112
    5 FORMAT(32HOPOLYNOMIAL REGRESSION OF DEGREE,I3)                      POLY0113
    6 FORMAT(12HO  INTERCEPT,E20.7)                                       POLY0114
    7 FORMAT(26HO  REGRESSION COEFFICIENTS/(6E20.7))                      POLY0115
    8 FORMAT(1HO/24X,24HANALYSIS OF VARIANCE FOR,I4,19H  DEGREE POLYNOMI  POLY0116
     1AL/)                                                                POLY0117
    9 FORMAT(1HO,5X,19HSOURCE OF VARIATION,7X,9HDEGREE OF,7X,6HSUM OF,9X  POLY0118
     1,4HMEAN,10X,1HF,9X,20HIMPROVEMENT IN TERMS/33X,7HFREEDOM,8X,7HSQUA  POLY0119
     2RES,7X,6HSQUARE,7X,5HVALUE,8X,17HOF SUM OF SQUARES)                 POLY0120
   10 FORMAT(20HO  DUE TO REGRESSION,12X,I6,F17.5,F14.5,F13.5,F20.5)      POLY0121
   11 FORMAT(32H   DEVIATION ABOUT REGRESSION   ,I6,F17.5,F14.5)          POLY0122
   12 FORMAT(8X,5HTOTAL,19X,I6,F17.5///)                                  POLY0123
   13 FORMAT(17HO  NO IMPROVEMENT)                                        POLY0124
   14 FORMAT(1HO//27X,18HTABLE OF RESIDUALS//16H OBSERVATION NO.,5X,7HX   POLY0125
     1VALUE,7X,7HY VALUE,7X,10HY ESTIMATE,7X,8HRESIDUAL/)                 POLY0126
   15 FORMAT(1HO,3X,I6,F18.5,F14.5,F17.5,F15.5)                           POLY0127
C                                                                         POLY0128
C     ..................................................................  POLY0129
C                                                                         POLY0130
C     READ PROBLEM PARAMETER CARD                                         POLY0131
      END=1000                                                            POLY0132
C                                                                         POLY0133
  100 READ (5,1) PR,PR1,N,M,NPLOT                                         POLY0134
C                                                                         POLY0135
C        PR....PROBLEM NUMBER (MAY BE ALPHAMERIC)                         POLY0136
C        PR1...PROBLEM NUMBER (CONTINUED)                                 POLY0137
C        N.....NUMBER OF OBSERVATIONS                                     POLY0138
C        M.....HIGHEST DEGREE POLYNOMIAL SPECIFIED                        POLY0139
C        NPLOT.OPTION CODE FOR PLOTTING                                   POLY0140
C              0  IF PLOT IS NOT DESIRED.                                 POLY0141
C              1  IF PLOT IS DESIRED.                                     POLY0142
C                                                                         POLY0143
                                                                          POLY0144
```

```
C      PRINT PROBLEM NUMBER AND N.                                          POLY0145
C                                                                           POLY0146
       WRITE (6,3) PR,PR1                                                   POLY0147
       WRITE (6,4) N                                                        POLY0148
C                                                                           POLY0149
C      READ INPUT DATA                                                      POLY0150
C                                                                           POLY0151
       L=N*N                                                                POLY0152
       DO 110 I=1,N                                                         POLY0153
       J=L+I                                                                POLY0154
C                                                                           POLY0155
C         X(I) IS THE INDEPENDENT VARIABLE, AND X(J) IS THE DEPENDENT       POLY0156
C         VARIABLE.                                                         POLY0157
C                                                                           POLY0158
  110  READ (5,2) X(I),X(J)                                                POLY0159
C                                                                           POLY0160
       CALL GDATA (N,M,X,XBAR,STD,D,SUMSQ)                                  POLY0161
C                                                                           POLY0162
       MM=M+1                                                               POLY0163
       SUM=0.0                                                              POLY0164
       NT=N-1                                                               POLY0165
C                                                                           POLY0166
       DO 200 I=1,M                                                         POLY0167
       ISAVE(I)=I                                                           POLY0168
C                                                                           POLY0169
C      FORM SUBSET OF CORRELATION COEFFICIENT MATRIX                        POLY0170
C                                                                           POLY0171
       CALL ORDER (MM,D,MM,I,ISAVE,DI,E)                                    POLY0172
C                                                                           POLY0173
C      INVERT THE SUBMATRIX OF CORRELATION COEFFICIENTS                     POLY0174
C                                                                           POLY0175
       CALL MINV (DI,I,DET,E,T)                                             POLY0176
C                                                                           POLY0177
       CALL MULTR (N,I,XBAR,STD,SUMSQ,DI,E,ISAVE,B,SB,T,ANS)                POLY0178
C                                                                           POLY0179
C      PRINT THE RESULT OF CALCULATION                                      POLY0180
```

```
C
      WRITE (6,5) I                                                    POLY0181
      IF(ANS(7)) 140,130,130                                           POLY0182
  130 SUMIP=ANS(4)-SUM                                                 POLY0183
      IF(SUMIP) 140, 140, 150                                          POLY0184
  140 WRITE (6,13)                                                     POLY0185
      GO TO 210                                                        POLY0186
  150 WRITE (6,6) ANS(1)                                               POLY0187
      WRITE (6,7) (B(J),J=1,I)                                         POLY0188
      WRITE (6,8) I                                                    POLY0189
      WRITE (6,9)                                                      POLY0190
      SUM=ANS(4)                                                       POLY0191
      WRITE (6,10) I,ANS(4),ANS(6),ANS(10),SUMIP                       POLY0192
      NI=ANS(8)                                                        POLY0193
      WRITE (6,11) NI,ANS(7),ANS(9)                                    POLY0194
      WRITE (6,12) NT,SUMSQ(MM)                                        POLY0195
C                                                                      POLY0196
C     SAVE COEFFICIENTS FOR CALCULATION OF Y ESTIMATES                 POLY0197
C                                                                      POLY0198
      COE(1)=ANS(1)                                                    POLY0199
      DO 160 J=1,I                                                     POLY0200
  160 COE(J+1)=B(J)                                                    POLY0201
      LA=I                                                             POLY0202
C                                                                      POLY0203
  200 CONTINUE                                                         POLY0204
C                                                                      POLY0205
C     TEST WHETHER PLOT IS DESIRED                                     POLY0206
C                                                                      POLY0207
  210 IF(NPLOT) 100, 100, 220                                          POLY0208
C                                                                      POLY0209
C        CALCULATE ESTIMATES                                           POLY0210
C                                                                      POLY0211
  220 NP3=N+N                                                          POLY0212
      DO 230 I=1,N                                                     POLY0213
      NP3=NP3+1                                                        POLY0214
      P(NP3)=COE(1)                                                    POLY0215
                                                                       POLY0216
```

```
      L=I                                                               POLY0217
      DO 230 J=1,LA                                                     POLY0218
      P(NP3)=P(NP3)+X(L)*COE(J+1)                                       POLY0219
  230 L=L+N                                                             POLY0220
C                                                                       POLY0221
C        COPY OBSERVED DATA                                             POLY0222
C                                                                       POLY0223
      N2=N                                                              POLY0224
      L=N*N                                                             POLY0225
      DO 240 I=1,N                                                      POLY0226
      P(I)=X(I)                                                         POLY0227
      N2=N2+1                                                           POLY0228
      L=L+1                                                             POLY0229
  240 P(N2)=X(L)                                                        POLY0230
C                                                                       POLY0231
C     PRINT TABLE OF RESIDUALS                                          POLY0232
C                                                                       POLY0233
      WRITE (6,3) PR,PR1                                                POLY0234
      WRITE (6,5) LA                                                    POLY0235
      WRITE (6,14)                                                      POLY0236
      NP2=N                                                             POLY0237
      NP3=N+N                                                           POLY0238
      DO 250 I=1,N                                                      POLY0239
      NP2=NP2+1                                                         POLY0240
      NP3=NP3+1                                                         POLY0241
      RESID=P(NP2)-P(NP3)                                               POLY0242
  250 WRITE (6,15) I,P(I),P(NP2),P(NP3),RESID                           POLY0243
C                                                                       POLY0244
      CALL PLOT (LA,P,N,3,C,1)                                          POLY0245
      GO TO 100                                                         POLY0246
C                                                                       POLY0247
      STOP                                                              POLY0248
 1000 CONTINUE                                                          POLY0249
      CALL EXIT                                                         POLY0250
      END                                                               POLY0251
C                                                                       POLY0252
```

```
C     ..................................................................   POLY0253
C                                                                          POLY0254
C        SUBROUTINE PLOT                                                   POLY0255
C                                                                          POLY0256
C        PURPOSE                                                           POLY0257
C           PLOT SEVERAL CROSS-VARIABLES VERSUS A BASE VARIABLE            POLY0258
C                                                                          POLY0259
C        USAGE                                                             POLY0260
C           CALL PLOT (NO,A,N,M,NL,NS)                                     POLY0261
C                                                                          POLY0262
C        DESCRIPTION OF PARAMETERS                                         POLY0263
C           NO - CHART NUMBER (3 DIGITS MAXIMUM)                           POLY0264
C           A  - MATRIX OF DATA TO BE PLOTTED. FIRST COLUMN REPRESENTS     POLY0265
C                BASE VARIABLE AND SUCCESSIVE COLUMNS ARE THE CROSS-       POLY0266
C                VARIABLES (MAXIMUM IS 9).                                 POLY0267
C           N  - NUMBER OF ROWS IN MATRIX A                                POLY0268
C           M  - NUMBER OF COLUMNS IN MATRIX A (EQUAL TO THE TOTAL         POLY0269
C                NUMBER OF VARIABLES). MAXIMUM IS 10.                      POLY0270
C           NL - NUMBER OF LINES IN THE PLOT. IF 0 IS SPECIFIED, 50        POLY0271
C                LINES ARE USED.                                           POLY0272
C           NS - CODE FOR SORTING THE BASE VARIABLE DATA IN ASCENDING      POLY0273
C                ORDER                                                     POLY0274
C                  0  SORTING IS NOT NECESSARY (ALREADY IN ASCENDING       POLY0275
C                     ORDER).                                              POLY0276
C                  1  SORTING IS NECESSARY.                                POLY0277
C                                                                          POLY0278
C        REMARKS                                                           POLY0279
C           NONE                                                           POLY0280
C                                                                          POLY0281
C        SUBROUTINES AND FUNCTION SUBPROGRAMS REQUIRED                     POLY0282
C           NONE                                                           POLY0283
C                                                                          POLY0284
C     ..................................................................   POLY0285
C                                                                          POLY0286
      SUBROUTINE PLOT(NO,A,N,M,NL,NS)                                      POLY0287
      DIMENSION OUT(101),YPR(11),ANG(9),A(1)                               POLY0288
```

```
      DATA BLANK/' '/                                                    POLY0289
      DATA ANG(1)/'1'/                                                   POLY0290
      DATA ANG(2)/'2'/                                                   POLY0291
      DATA ANG(3)/'3'/                                                   POLY0292
      DATA ANG(4)/'4'/                                                   POLY0293
      DATA ANG(5)/'5'/                                                   POLY0294
      DATA ANG(6)/'6'/                                                   POLY0295
      DATA ANG(7)/'7'/                                                   POLY0296
      DATA ANG(8)/'8'/                                                   POLY0297
      DATA ANG(9)/'9'/                                                   POLY0298
C                                                                        POLY0299
    1 FORMAT(1H1,60X,7H CHART ,I3,//)                                    POLY0300
    2 FORMAT(1H ,F11.4,5X,101A1)                                         POLY0301
    3 FORMAT(1H )                                                        POLY0302
    4 FORMAT(10H 123456789)                                              POLY0303
    5 FORMAT(10A1)                                                       POLY0304
    7 FORMAT(1H ,16X,101H.         .         .         .         .       POLY0305
     1   .         .         .         .         .         .)            POLY0306
    8 FORMAT(1HO,9X,11F10.4)                                             POLY0307
C                                                                        POLY0308
C     ..................................................................POLY0309
C                                                                        POLY0310
      NLL=NL                                                             POLY0311
C                                                                        POLY0312
      IF(NS) 16, 16, 10                                                  POLY0313
C                                                                        POLY0314
C        SORT BASE VARIABLE DATA IN ASCENDING ORDER                      POLY0315
C                                                                        POLY0316
   10 DO 15 I=1,N                                                        POLY0317
      DO 14 J=I,N                                                        POLY0318
      IF(A(I)-A(J)) 14, 14, 11                                           POLY0319
   11 L=I-N                                                              POLY0320
      LL=J-N                                                             POLY0321
      DO 12 K=1,M                                                        POLY0322
      L=L+N                                                              POLY0323
      LL=LL+N                                                            POLY0324
```

```
      F=A(L)                                                            POLY0325
      A(L)=A(LL)                                                        POLY0326
   12 A(LL)=F                                                           POLY0327
   14 CONTINUE                                                          POLY0328
   15 CONTINUE                                                          POLY0329
C                                                                       POLY0330
C        TEST NLL                                                       POLY0331
C                                                                       POLY0332
   16 IF(NLL) 20, 18, 20                                                POLY0333
   18 NLL=50                                                            POLY0334
C                                                                       POLY0335
C        PRINT TITLE                                                    POLY0336
C                                                                       POLY0337
   20 WRITE(6,1)NO                                                      POLY0338
C                                                                       POLY0339
C        DEVELOP BLANK AND DIGITS FOR PRINTING                          POLY0340
C                                                                       POLY0341
C                                                                       POLY0342
C        FIND SCALE FOR BASE VARIABLE                                   POLY0343
C                                                                       POLY0344
      XSCAL=(A(N)-A(1))/(FLOAT(NLL-1))                                  POLY0345
C                                                                       POLY0346
C        FIND SCALE FOR CROSS-VARIABLES                                 POLY0347
C                                                                       POLY0348
      M1=N+1                                                            POLY0349
      YMIN=A(M1)                                                        POLY0350
      YMAX=YMIN                                                         POLY0351
      M2=M*N                                                            POLY0352
      DO 40 J=M1,M2                                                     POLY0353
      IF(A(J)-YMIN) 28,26,26                                            POLY0354
   26 IF(A(J)-YMAX) 40,40,30                                            POLY0355
   28 YMIN=A(J)                                                         POLY0356
      GO TO 40                                                          POLY0357
   30 YMAX=A(J)                                                         POLY0358
   40 CONTINUE                                                          POLY0359
      YSCAL=(YMAX-YMIN)/100.0                                           POLY0360
```

```
C                                                                       POLY0361
C         FIND BASE VARIABLE PRINT POSITION                             POLY0362
C                                                                       POLY0363
      XB=A(1)                                                           POLY0364
      L=1                                                               POLY0365
      MY=M-1                                                            POLY0366
      I=1                                                               POLY0367
   45 F=I-1                                                             POLY0368
      XPR=XB+F*XSCAL                                                    POLY0369
      IF(A(L)-XPR) 50,50,70                                             POLY0370
C                                                                       POLY0371
C         FIND CROSS-VARIABLES                                          POLY0372
C                                                                       POLY0373
   50 DO 55 IX=1,101                                                    POLY0374
   55 OUT(IX)=BLANK                                                     POLY0375
      DO 60 J=1,MY                                                      POLY0376
      LL=L+J*N                                                          POLY0377
      JP=((A(LL)-YMIN)/YSCAL)+1.0                                       POLY0378
      OUT(JP)=ANG(J)                                                    POLY0379
   60 CONTINUE                                                          POLY0380
C                                                                       POLY0381
C         PRINT LINE AND CLEAR, OR SKIP                                 POLY0382
C                                                                       POLY0383
      WRITE(6,2)XPR,(OUT(IZ),IZ=1,101)                                  POLY0384
      L=L+1                                                             POLY0385
      GO TO 80                                                          POLY0386
   70 WRITE(6,3)                                                        POLY0387
   80 I=I+1                                                             POLY0388
      IF(I-NLL) 45, 84, 86                                              POLY0389
   84 XPR=A(N)                                                          POLY0390
      GO TO 50                                                          POLY0391
C                                                                       POLY0392
C         PRINT CROSS-VARIABLES NUMBERS                                 POLY0393
C                                                                       POLY0394
   86 WRITE(6,7)                                                        POLY0395
      YPR(1)=YMIN                                                       POLY0396
```

```
      DO 90 KN=1,9                                                    POLY0397
   90 YPR(KN+1)=YPR(KN)+YSCAL*10.0                                    POLY0398
      YPR(11)=YMAX                                                    POLY0399
      WRITE(6,8)(YPR(IP),IP=1,11)                                     POLY0400
      RETURN                                                          POLY0401
      END                                                             POLY0402
```

```
C                                                                       FFBZ0001
C                                                                       FFBZ0002
C                                                                       FFBZ0003
C     ************************************************************      FFBZ0004
C     *                                                          *      FFBZ0005
C     *                                                          *      FFBZ0006
C     *            FISSION FRAGMENTS IN BENZENE                  *      FFBZ0007
C     *                                                          *      FFBZ0008
C     *                                                          *      FFBZ0009
C     *                   ENERGY BALANCE METHOD                  *      FFBZ0010
C     *                                                          *      FFBZ0011
C     *                                                          *      FFBZ0012
C     ************************************************************      FFBZ0013
C                                                                       FFBZ0014
C                                                                       FFBZ0015
C                                                                       FFBZ0016
C                                                                       FFBZ0017
C                                                                       FFBZ0018
C                                                                       FFBZ0019
C                                                                       FFBZ0020
C                                                                       FFBZ0021
C                                                                       FFBZ0022
      IMPLICIT REAL*8 (A-H,C-Z)                                         FFBZ0023
      EXP(X)=DEXP(X)                                                    FFBZ0024
      ALOG(X)=DLOG(X)                                                   FFBZ0025
      SQRT(X)=DSQRT(X)                                                  FFBZ0026
      ABS(X)=DABS(X)                                                    FFBZ0027
C                                                                       FFBZ0028
      DIMENSION PLIQ(30),TSAT(30)                                       FFBZ0029
      DIMENSION A(30)                                                   FFBZ0030
  500 FORMAT(F10.4)                                                     FFBZ0031
  501 FORMAT(I3/(10X,2F10.2))                                           FFBZ0032
  503 FORMAT(1H1,30X,' THE SUPERHEAT LIMIT FOR  BENZENE  EXPOSED        FFBZ0033
     1 TO FISSION FRAGMENTS '////)                                      FFBZ0034
  504 FORMAT(1H0,119H A PKO GT EPKA DELTI  ALPHA  PRESS   SAT    ENER  A FFBZ0035
     1VG ENER   J   XNI  REL SIZE   GAS    VAPOR   DEL T   DEL T    SUP FFBZ0036
```

```
     1  )                                                                    FFBZ0037
  505 FORMAT(1H ,119H        RATE      TIME          LIQUID  TEMP   LEVEL C  FFBZ0038
     1EP RATE            INIT EMBR  PRESS   PRESS    EQA48   E.B.M.  TEMP    FFBZ0039
     1  )                                                                    FFBZ0040
  506 FORMAT(1HO,119H      NC/SEC      SEC          LBF/FT2 DEG F     MEV  M  FFBZ0041
     1EV/CM                        LBF/FT2 LBF/FT2  DEG F   DEG F   DEG F   FFBZ0042
     1  )                                                                    FFBZ0043
  508 FORMAT(1H ,F5.2,D6.3 ,D8.1,F6.3,F8.1,F8.3,F9.5,D10.3,I5,F5.1,F10.8     FFBZ0044
     1, F7.1,F8.1,8X,F8.3,F8.3)                                             FFBZ0045
  600 FORMAT(1HO,29HTHE VALUE OF THE CONSTANT IS ,F10.4//)                   FFBZ0046
  999 FORMAT(I3/(F10.3))                                                     FFBZ0047
  601 FORMAT(8H EPKA = ,F10.4///)                                            FFBZ0048
C                                                                            FFBZ0049
C      EPKA IS THE INITIAL ENERGY OF THE FISSION FRAGMENT                    FFBZ0050
C                                                                            FFBZ0051
      EPKA = 95.0                                                            FFBZ0052
C                                                                            FFBZ0053
    1 READ(5,501)N,(PLIQ(L),TSAT(L),L=1,N)                                   FFBZ0054
      READ(5,999)K,(A(M),M=1,K)                                              FFBZ0055
    3 WRITE(6,503)                                                           FFBZ0056
    4 WRITE(6,504)                                                           FFBZ0057
    5 WRITE(6,505)                                                           FFBZ0058
    6 WRITE(6,506)                                                           FFBZ0059
 1000 READ(5,500)CONST                                                       FFBZ0060
      WRITE(6,600)CONST                                                      FFBZ0061
      WRITE(6,601)EPKA                                                       FFBZ0062
      DO 110 M=1,K,1                                                         FFBZ0063
    8 DO 109 L=1,N,1                                                         FFBZ0064
   10 DELTF=10.0                                                             FFBZ0065
   15 TF=TSAT(L)+DELTF                                                       FFBZ0066
   23 CONTINUE                                                               FFBZ0067
C                                                                            FFBZ0068
C          PHYSICAL PROPERTIES OF BENZENE                                    FFBZ0069
C                                                                            FFBZ0070
   25 PVA = 85.143 - 0.9599*TF + 0.003178*TF*TF                              FFBZ0071
   26 PVPSF = PVA*144.0                                                      FFBZ0072
```

```
   27 SIGMA = 2.268E-3 - 4.61CE-6*TF                                    FFBZ0073
   46 ROWL = 1.C/(C.01831 + C.15524E-5*TF + 0.3444E-7*TF*TF)            FFBZ0074
   55 ROWV = 1.C/(26.8282 - C.1614*TF + 0.24878E-3*TF*TF)               FFBZ0075
   56 PPC = 196.119 - 0.12169*TF - 0.1734E-3*TF*TF                      FFBZ0076
   57 BA=(PVPSF-PLIQ(L))/PVPSF                                          FFBZ0077
C                                                                       FFBZ0078
      E = 10.0                                                          FFBZ0079
C                                                                       FFBZ0080
      ROWL=CONST*ROWL                                                   FFBZ0081
C                                                                       FFBZ0082
C                                                                       FFBZ0083
C       BENZENE PARAMETERS FOR FISSION FRAGMENTS                        FFBZ0084
C                                                                       FFBZ0085
C                                                                       FFBZ0086
C                                                                       FFBZ0087
C                                                                       FFBZ0088
 C158 COEF = (3.4212E04)*ROWL*A(M)*SIGMA/(BA*PVPSF)                     FFBZ0089
 C258 CCC = EPKA*EPKA - COEF*EPKA*(ALOG(0.0622*EPKA)-1.0)               FFBZ0090
 3258 CDD = 2.C*EPKA + 3.777*COEF                                       FFBZ0091
 C358 BBB = CDD - COEF*ALOG(E)                                          FFBZ0092
C                                                                       FFBZ0093
 C458 EC=E-(E*E-BBB*E+CCC)/(2.0*E-BBB)                                  FFBZ0094
 0558 IF((ABS(E-EC)-0.0010).LT.0.0) GO TO 9858                          FFBZ0095
      E = ( E + EC )* 0.5                                               FFBZ0096
      IF ( E.LE.0.C) E = 1.C                                            FFBZ0097
 C758 GO TO 0358                                                        FFBZ0098
 9858 IF((EC-10.0C).LT.0.C) GO TO 96                                    FFBZ0099
      IF((EC-EPKA).GE.0.0) GO TO 93                                     FFBZ0100
      DEDS = (EPKA-E)*BA*PVPSF/(60.96*A(M)*SIGMA)                       FFBZC101
      RCRIT=2.C*SIGMA/(BA*PVPSF)                                        FFBZ0102
   62 GATE=3.700221E-19*A(M)*(TF+459.69)*DEDS/SIGMA**2                  FFBZ0103
      ART=(2.C*GATE*(PVPSF-PLIQ(L))-1.0)**2-4.C*GATE**2*(PVPSF-PLIQ(L))*FFBZ0104
     1*2                                                                FFBZ0105
      IF(ART.LT.0.C) GO TO 93                                           FFBZ0106
   63 PGPSF =(1.0-2.0*GATE*(PVPSF -PLIC(L))-SQRT(ART))/(2.0*GATE)       FFBZ0107
   64 E=(PVPSF +PGPSF -PLIC(L))/PVPSF                                   FFBZ0108
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```
0164  IF((B-BA).LT.0.00001) GO TO 464                                          FFBZ0109
      BA = B                                                                   FFBZ0110
0364  GO TO 0158                                                               FFBZ0111
 464  GO TO 103                                                                FFBZ0112
 103  EQA53=0.8010E-12*A(M)*DEDS-RCRIT*RCRIT*RCWV*HFG*778.26-RCRIT*SIGMA       FFBZ0113
 104  IF(EQA53-0.0)9696,106,9393                                               FFBZ0114
9696  IF(ABS(EQA53).LE.1.0E-16) GO TO 106                                      FFBZ0115
      GO TO 96                                                                 FFBZ0116
9393  IF(ABS(EQA53).LE.1.0E-16) GO TO 106                                      FFBZ0117
      GO TO 93                                                                 FFBZ0118
  93  TF=TF-DELTF                                                              FFBZ0119
  94  DELTF=0.1*DELTF                                                          FFBZ0120
  95  IF(DELTF.LT.0.009) GO TO 106                                             FFBZ0121
  96  TF=TF+DELTF                                                              FFBZ0122
  97  GO TO 23                                                                 FFBZ0123
 106  DELT3=TF-TSAT(L)                                                         FFBZ0124
      PKO2RT=0.0                                                               FFBZ0125
      XI=0.0                                                                   FFBZ0126
      DELTI=0.0                                                                FFBZ0127
      ALPHA=0.0                                                                FFBZ0128
      J=0                                                                      FFBZ0129
      XN1=0.0                                                                  FFBZ0130
 107  WRITE(6,508)A(M),PKO2RT,DELTI,ALPHA,PLIQ(L),TSAT(L),E    ,DEDS   ,J,    FFBZ0131
     1XNI,XI,PGPSF,PVPSF,DELT3,TF                                              FFBZ0132
 109  CONTINUE                                                                 FFBZ0133
   7  WRITE(6,900)                                                             FFBZ0134
 900  FORMAT(//45X,22H DIFFERENT VALUE OF A ,//)                               FFBZ0135
 110  CONTINUE                                                                 FFBZ0136
      GO TO 1000                                                               FFBZ0137
      END                                                                      FFBZ0138
```

```
C                                                                       FNBZ0001
C                                                                       FNBZ0002
C                                                                       FNBZ0003
C     ************************************************************      FNBZ0004
C     *                                                          *      FNBZ0005
C     *                                                          *      FNBZ0006
C     *               FAST NEUTRONS IN BENZENE                   *      FNBZ0007
C     *                                                          *      FNBZ0008
C     *                                                          *      FNBZ0009
C     *                    ENERGY BALANCE METHOD                 *      FNBZ0010
C     *                                                          *      FNBZ0011
C     *                                                          *      FNBZ0012
C     ************************************************************      FNBZ0013
C                                                                       FNBZ0014
C                                                                       FNBZ0015
C                                                                       FNBZ0016
      IMPLICIT REAL*8 (A-H,C-Z)                                         FNBZ0017
      EXP(X)=DEXP(X)                                                    FNBZ0018
      ALOG(X)=DLOG(X)                                                   FNBZ0019
      SQRT(X)=DSQRT(X)                                                  FNBZ0020
      ABS(X)=DABS(X)                                                    FNBZ0021
C                                                                       FNBZ0022
      DIMENSION PLIQ(30),TSAT(30)                                       FNBZ0023
      DIMENSION A(30)                                                   FNBZ0024
  500 FORMAT(F10.4)                                                     FNBZ0025
  501 FORMAT(I3/(10X,2F10.2))                                           FNBZ0026
  503 FORMAT(1H1,30X,' THE SUPERHEAT LIMIT FOR      BENZENE     EXPOSED FNBZ0027
     1 TO  FAST NEUTRONS     '////)                                     FNBZ0028
  504 FORMAT(1HO,119H A PKO GT EPKA DELTI  ALPHA  PRESS   SAT    ENER  A FNBZ0029
     1VG ENER   J   XNI  REL SIZE   GAS    VAPOR   DEL T   DEL T    SUP FNBZ0030
     1  )                                                              FNBZ0031
  505 FORMAT(1H ,119H       RATE     TIME         LIQUID  TEMP   LEVEL D FNBZ0032
     1EP RATE          INIT EMBR  PRESS   PRESS   EQA48   E.B.M.  TEMP  FNBZ0033
     1  )                                                              FNBZ0034
  506 FORMAT(1HO,119H     NC/SEC     SEC         LBF/FT2 DEG F    MEV  M FNBZ0035
     1EV/CM                    LBF/FT2 LBF/FT2  DEG F   DEG F   DEG F  FNBZ0036
```

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     1  )                                                                  FNBZ0037
   508 FORMAT(1H ,F5.2,D6.3 ,D8.1,F6.3,F8.1,F8.3,F9.5,D10.3,I5,F5.1,F10.8  FNBZ0038
     1, F7.1,F8.1,8X,F8.3,F8.3)                                           FNBZ0039
   600 FORMAT(1H0,29HTHE VALLE OF THE CONSTANT IS ,F10.4//)               FNBZ0040
   999 FORMAT(I3/(F10.3))                                                 FNBZ0041
   601 FORMAT(8H EPKA = ,F10.4///)                                        FNBZ0042
C                                                                         FNBZ0043
C      EPKA IS THE ENERGY OF THE PKOA                                     FNBZ0044
C                                                                         FNBZ0045
       EPKA = 2.12                                                        FNBZ0046
C                                                                         FNBZ0047
     1 READ(5,501)N,(PLIQ(L),TSAT(L),L=1,N)                               FNBZ0048
       READ(5,999)K,(A(M),M=1,K)                                          FNBZ0049
     3 WRITE(6,503)                                                       FNBZ0050
     4 WRITE(6,504)                                                       FNBZ0051
     5 WRITE(6,505)                                                       FNBZ0052
     6 WRITE(6,506)                                                       FNBZ0053
  1000 READ(5,500)CONST                                                   FNBZ0054
       WRITE(6,600)CONST                                                  FNBZ0055
       WRITE(6,601)EPKA                                                   FNBZ0056
       DO 110 M=1,K,1                                                     FNBZ0057
     8 DO 109 L=1,N,1                                                     FNBZ0058
    10 DELTF=10.0                                                         FNBZ0059
    15 TF=TSAT(L)+DELTF                                                   FNBZ0060
    23 CONTINUE                                                           FNBZ0061
C                                                                         FNBZ0062
C           PHYSICAL PROPERTIES OF BENZENE                                FNBZ0063
C                                                                         FNBZ0064
    25 PVA = 85.143 - 0.9599*TF + 0.003178*TF*TF                          FNBZ0065
    26 PVPSF = PVA*144.0                                                  FNBZ0066
    27 SIGMA = 2.268E-3 - 4.610E-6*TF                                     FNBZ0067
    46 ROWL = 1.0/(0.01831 + 0.15524E-5*TF + 0.3444E-7*TF*TF)             FNBZ0068
    55 ROWV = 1.0/(26.8282 - 0.1614*TF + 0.24878E-3*TF*TF)                FNBZ0069
    56 HFG = 196.119 - 0.12169*TF - 0.1734E-3*TF*TF                       FNBZ0070
    57 BA=(PVPSF-PLIQ(L))/PVPSF                                           FNBZ0071
C                                                                         FNBZ0072
```

```
      E = 1.50                                                                  FNBZ0073
C                                                                               FNBZ0074
      ROWL=CONST*RCWL                                                           FNBZ0075
C                                                                               FNBZ0076
C                                                                               FNBZ0077
 0158 COEF = (1.00E04*ROWL*A(M)*SIGMA)/(BA*PVPSF)                               FNBZ0078
 0258 CCC = EPKA*EPKA - COEF*EPKA*(ALOG(0.9450*EPKA)-1.0)                       FNBZ0079
 3258 DDD = 1.0565*COEF + 2.0*EPKA                                              FNBZ0080
 0358 BBB = DDD - COEF*ALOG(E)                                                  FNBZ0081
C                                                                               FNBZ0082
 0458 EC=E-(E*E-BBB*E+CCC)/(2.0*E-BBB)                                          FNBZ0083
 0558 IF((ABS(E-EC)-0.0030).LT.0.0) GO TO 9858                                  FNBZ0084
      E = ( E + EC )* 0.5                                                       FNBZ0085
      IF ( E.LE.0.C) E = 1.C                                                    FNBZ0086
 0758 GO TO 0358                                                                FNBZ0087
 9858 IF((EC-10.00).LT.0.0) GO TO 96                                            FNBZ0088
      IF((EC-EPKA).GE.0.0) GO TO 93                                             FNBZ0089
      DEDS = (EPKA-E)*BA*PVPSF/(60.96*A(M)*SIGMA)                               FNBZ0090
      RCRIT=2.0*SIGMA/(BA*PVPSF)                                                FNBZ0091
   62 GATE=3.700221E-19*A(M)*(TF+459.69)*DEDS/SIGMA**2                          FNBZ0092
      ART=(2.0*GATE*(PVPSF-PLIQ(L))-1.0)**2-4.0*GATE**2*(PVPSF-PLIQ(L))*        FNBZ0093
     1*2                                                                        FNBZ0094
      IF(ART.LT.0.0) GO TO 93                                                   FNBZ0095
   63 PGPSF =(1.0-2.0*GATE*(PVPSF -PLIQ(L))-SQRT(ART))/(2.0*GATE)               FNBZ0096
   64 B=(PVPSF +PGPSF -PLIQ(L))/PVPSF                                           FNBZ0097
 0164 IF((B-BA).LT.0.00001) GO TO 464                                           FNBZ0098
      BA = B                                                                    FNBZ0099
 0364 GO TO 0158                                                                FNBZ0100
  464 GO TC 103                                                                 FNBZ0101
  103 EQA53=0.8010E-12*A(M)*DEDS-RCRIT*RCRIT*RCWV*HFG*778.26-RCRIT*SIGMA        FNBZ0102
  104 IF(EQA53-0.0)9696,106,9393                                                FNBZ0103
 9696 IF(ABS(EQA53).LE.1.0E-16) GO TO 106                                       FNBZ0104
      GO TO 96                                                                  FNBZ0105
 9393 IF(ABS(EQA53).LE.1.0E-16) GO TO 106                                       FNBZ0106
      GO TC 93                                                                  FNBZ0107
   93 TF=TF-DELTF                                                               FNBZ0108
```

```
 94 DELTF=0.1*DELTF                                                        FNBZ0109
 95 IF(DELTF.LT.0.009) GO TO 106                                           FNBZ0110
 96 TF=TF+DELTF                                                            FNBZ0111
 97 GO TO 23                                                               FNBZ0112
106 DELT3=TF-TSAT(L)                                                       FNBZ0113
    PKO2RT=0.0                                                             FNBZ0114
    XI=0.0                                                                 FNBZ0115
    DELTI=0.0                                                              FNBZ0116
    ALPHA=0.0                                                              FNBZ0117
    J=0                                                                    FNBZ0118
    XN1=0.0                                                                FNBZ0119
107 WRITE(6,508)A(M),PKO2RT,DELTI,ALPHA,PLIQ(L),TSAT(L),E    ,DEDS   ,J,   FNBZ0120
   1XNI,XI,PGPSF,PVPSF,DELT3,TF                                            FNBZ0121
109 CONTINUE                                                               FNBZ0122
  7 WRITE(6,900)                                                           FNBZ0123
900 FORMAT(//45X,22H DIFFERENT VALUE OF A ,//)                             FNBZ0124
110 CONTINUE                                                               FNBZ0125
    GO TO 1000                                                             FNBZ0126
    END                                                                    FNBZ0127
```

Appendix D

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