Prediction of the minimum neutron energy to nucleate vapor bubbles in superheated liquids

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Calculated values of the stopping power of ions in superheated liquid have been used in predicting the minimum neutron energy that will nucleate vapor bubbles in a superheated liquid. Such predictions are useful in understanding the behavior of a new class of neutron detectors based on superheated halocarbon or hydrocarbon drops suspended in aqueous gel.

INTRODUCTION

It is known that a liquid can be raised to a temperature higher than its boiling point without vaporizing. This metastable state of liquid known as the superheated state is normally short lived due to the presence of heterogeneous nucleation sites (e.g., air bubbles, solid impurities, or gas pockets at a liquid-solid interface such as a container surface). The bubble chamber discovered by Glaser[1] in 1952 makes use of the liquid's ability to remain momentarily superheated so that an elementary particle's path can be tracked by the bubbles it nucleates as it traverses the liquid. Apfel's superheated drop detector (SDD) is a collection of drops of superheated liquid suspended in another liquid, ordinarily a viscous immiscible gel.[2,3] It uses the same basic principle as that of bubble chambers: namely, the initiation of vapor bubbles by ions in superheated liquids. If the bubbles reach a size that makes them thermodynamically unstable (the "critical" size), the bubbles grow to observable size through the evaporation of the superheated liquid. Viscous and interfacial forces may prevent this size from being reached. The critical radius is given by:

$$R_c = 2\gamma(T)/\Delta P \ , \tag{1}$$

where $\gamma(T)$ is the surface tension at a temperature T, and $\Delta P = P_v(T) + P_g - P_0$ with P_v the pressure of the vapor in the cavity, P_g the partial pressure associated with the noncondensible dissolved gas, and P_0 the externally applied pressure. Obviously, for larger ΔP, R_c is smaller and the heat required for drop vaporization is less.

Seitz originally suggested that ions deposit energy locally via a "thermal spike" which produces critically sized vapor nuclei.[4] The energy deposited along that part of the ion's range corresponding to about twice the critical radius will contribute significantly to bubble formation. The energy deposited by a particular ion in a given distance of sample (one critical diameter in our case) can be

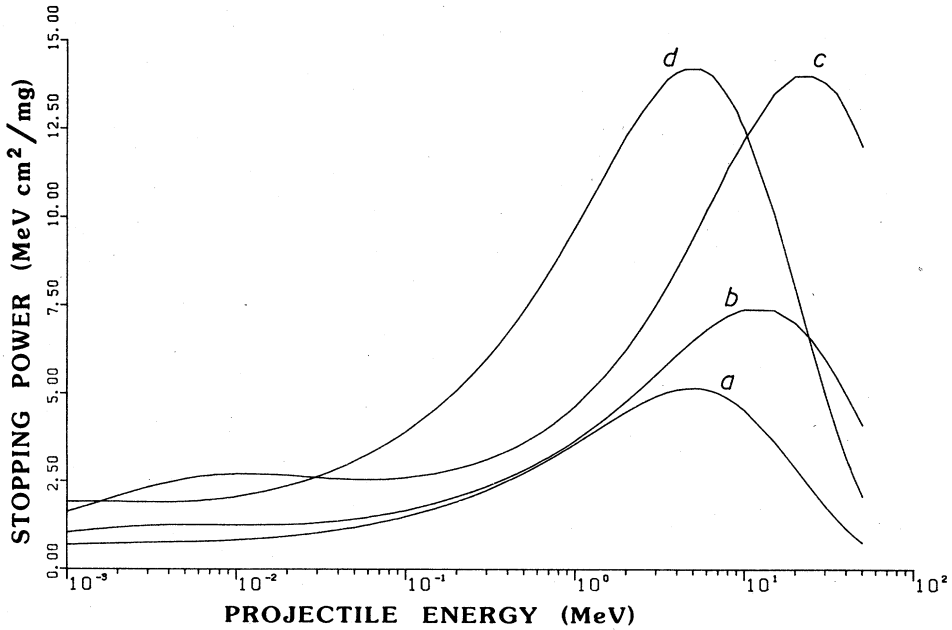

FIG. 1. Stopping powers of relevant projectiles in liquids of SDD interest. (a) carbon in Freon-12; (b) fluorine in Freon-12; (c) chlorine in Freon-12; (d) carbon in isobutane.

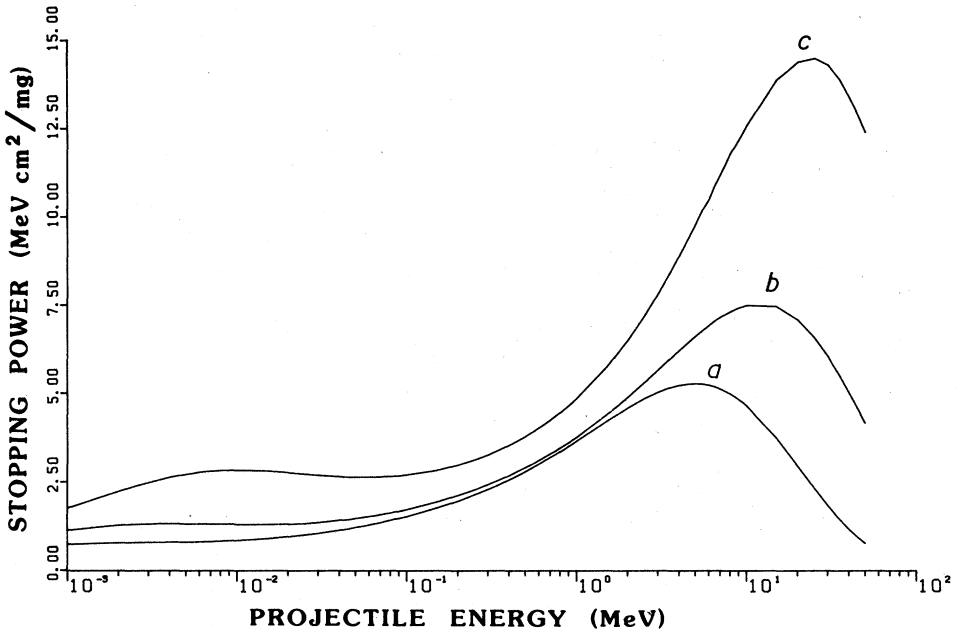

FIG. 2. Stopping powers of different projectiles in Freon-114. (a) carbon; (b) fluorine; (c) chlorine.

computed from the conventional stopping power (dE/dx) of that ion in that sample. As different ions expend different amounts of energy in a given sample, determination of the threshold energy of vapor nucleation of a given liquid depends on which of the relevant ions expends most of its energy. When neutrons have energy E_n, the maximum amount of energy that the nucleus of atomic weight A can receive from the neutron is

$$4AE_n/(A+1)^2 , \tag{2}$$

via the elastic head-on collision. The nucleus is "shot out" from its electrons and shuttles through the liquid depositing its energy until electron collisions and charge capture bring the ion to rest.

As an example, when neutrons of a given energy hit a drop of refrigerant 12 (CCl_2F_2; DuPont trade name Freon-12), it is possible that the different nuclei of the liquid (namely C, Cl, F) will receive different amounts of energy. The one which will play the major role in vapor nucleation is determined by the ion which deposits the most energy in a given critical diameter. Therefore, knowledge of (dE/dx) of relevant ions in relevant liquids plays a central role in predicting the threshold neutron energy to trigger a given liquid.

For neutron dosimetry applications the stopping power of ions is usually in an intermediate range between the monotonically increasing behavior at low ion energy and the monotonically decreasing behavior at high energy. [One exception is for sufficiently massive ions of sufficiently low energy; for these ions there is a nonnegligible contribution to the stopping power due to the interaction of the ion with nuclei of the target (nuclear stopping power).] Consequently, modeling the physics of the intermediate range is difficult, and there are no simple analytic formulas for this range which allow for the prediction of stopping power for different ion-target combinations.

In addition to the well-known tables of Northcliffe and Schilling,[5] few formulas have been derived that will give predictions that correspond to experimental data if certain other experimental data are provided and/or if certain adjustable parameters are employed.[6–11] In what follows we use a recently derived semiempirical formula[12] to calculate the stopping power, dE/dx, for ions appropriate to our neutron dosimetry work for energies above about 0.2 MeV. For ions below this energy we calculate the total stopping power as the sum of the electronic stopping power, using Firsov's formula,[13] and the nuclear stopping power, using Ziegler's formula,[14] which is claimed to be more accurate than Lindhard, Sharff, and Schiott (LSS) nuclear stopping power.[15] We present in Figs. 1 and 2 dE/dx values of relevant ions in three different SDD liquids [Freon-12, CCl_2F_2, (bp)~ -30 °C; Freon-114, $C_2Cl_2F_4$, bp~ 4 °C, and isobutane, C_4H_{10}, bp~ -11 °C]. Apart from problems in developing a correct theory for vapor nucleation, there exists a scarcity of experimental results for nucleation in different liquids over a wide range of nuclear, fluid, and thermal parameters.[2,4] The present work is an attempt to fill this data void to a certain extent using different liquids under varying conditions.

MEASUREMENT, INTERPRETATION, AND PREDICTION OF THRESHOLD ENERGY

In experiments with carefully controlled temperature, using procedures described in Ref. 2, the threshold neutron energies to trigger superheated drops of Freon-114 at two different temperatures are found to be 4.1 MeV at

TABLE I. Certain observed and other physical parameters for Freon-114 ($C_2Cl_2F_4$) at two different temperatures.

Items	Temperature 26.5 °C	41.3 °C
1. Observed threshold neutron energy	4.1 MeV	0.46 MeV
2. Maximum energy imparted to:		
carbon	1.16 MeV	0.13 MeV
fluorine	0.78 MeV	0.087 MeV
chlorine	0.45 MeV	0.05 MeV
3. Surface tension (dyn/cm)	11.85	10.38
4. ΔP (atm); 1 atm $=10^5$ Pa	1.28	2.60
5. Critical radius ($\times 10^{-6}$ cm)	18.51	7.98
6. (dE/dx) in MeV cm^2/mg corresponding to ion in item 2 that deposits the maximum energy	3.75 (carbon)	1.56 (chlorine)
7. Energy, E_c, deposited in critical diameter in keV	204[a]	50[b]
8. Gibbs energy, W, in keV, from Eq. (3)	10.64	1.73
9. $\eta=\frac{W}{E_c}$	0.052	0.035
10. Inhibition temperature ~ 13 °C (based on a chlorine ion projectile)		

[a] $E_c=(dE/dx)\times 2R_c$

[b] This is the entire chlorine ion energy because the range of this ion is less than $2R_c$.

26.5 °C and 0.46 MeV at 41.3 °C and for isobutane 0.84 MeV at 24 °C and 0.27 MeV at 40 °C. We present in Table I for the case of Freon-114 the corresponding maximum energies that the carbon, fluorine, and chlorine nuclei may get as calculated using Eq. (2). In Table II, we present the corresponding maximum energies the carbon and hydrogen nuclei in isobutane may get. In any case, it has been found from the corresponding (dE/dx) curves that for these liquids it is the carbon ion that deposits the most energy in a critical diameter except for the single case of Freon-114 at 41.3 °C for which the chlorine ion deposits slightly more energy in a critical diameter than the carbon or fluorine ion. The amount of energy deposited by the different ions in the critical diameters at two different temperatures along with some other relevant physical parameters for Freon-114 and isobutane are presented in Tables I and II, respectively.

In addition to presenting in the tables the approximate energy deposited along a critical diameter, which we call E_c, we also give the reversible thermodynamic work to form a vapor bubble of critical size

$$W=16\pi\gamma^3(T)/3(\Delta P)^2 \tag{3}$$

which was originally derived by Gibbs.[16] The ratio

TABLE II. Certain observed and other physical parameters for isobutane (C_4H_{10}) at two different temperatures.

Items	Temperature 24 °C	40 °C
1. Observed threshold neutron energy	0.84 MeV	0.27 MeV
2. Maximum energy imparted to:		
carbon	0.24 MeV	0.077 MeV
hydrogen	0.84 MeV	0.27 MeV
3. Surface tension (dyn/cm)	11.08	9.48
4. ΔP (atm); 1 atm $=10^5$ Pa	2.36	4.25
5. Critical radius ($\times 10^{-6}$ cm)	9.39	4.46
6. (dE/dX) in MeV cm^2/mg corresponding to maximum carbon energy in item 2, above	5.39	3.03
7. Energy, E_c, deposited in critical diameter in keV	56.7[a]	15.1[a]
8. Gibbs energy, W, in keV from Eq. (3)	2.56	0.49
9. $\eta=W/E_c$	0.045	0.032
10. Inhibition temperature ~ 9 °C (based on a carbon ion projectile)		

[a] $E_c=(dE/dx)\times 2R_c$

$\eta \equiv W/E_c$ is given by:

$$\eta \equiv \frac{W}{E_c} = \frac{4\pi\gamma^2}{3(\Delta P)(dE/dx)} \tag{4}$$

for the case when E_c is estimated by $(dE/dx)\times 2R_c$. η is a rough measure of the efficiency (or "inefficiency") of the actual nucleation process as compared to the ideal reversible thermodynamic one. For our two liquids it varies from about 3% to 5%.

We also decided to apply the same analysis to the data of Greenspan and Tschiegg,[17] who measured the temperature dependence of the acoustic cavitation threshold for liquids exposed to a Pu-Be neutron source (maximum neutron energy is approximately 10 MeV). In Figure 3 the solid symbols represent η calculated using Eq. (4) for each of three liquids. η is plotted against $\Delta P/P_c$, where for the acoustic case $\Delta P = P_v(T) + P_A - P_0$. Here P_A is the peak acoustic pressure and P_c is the critical pressure for the sample. Also plotted with open symbols are the results for our superheated samples, as given in Tables I and II. It is clear from Fig. 3 that η varies only from 3% to 5% even though the conditions of irradiation and of nucleation (i.e., superheat versus acoustic waves) vary greatly. We conclude that the physics of the nucleation process is the same for both sets of experiments.

It is fair to ask why the process of radiation-induced nucleation is such an inefficient process. A clue to the answer may be found in the preliminary work of Apfel, Chu, and Mengel,[18] who have been considering a dynamic theory following on the less quantitative analysis of Seitz.[4] In the more recent work, Apfel, Chu, and Mengel discuss theoretically the dynamic factors resulting from the sudden deposit of heat along a line in the superheated liquid. The first phase involves the generation of a strong shock wave resulting from the heating of a small region to temperatures and pressures far beyond their critical values. When, at some time later, the hot, high-pressure region has expanded sufficiently so that at some radius the critical parameters are achieved, then an interface separating liquid and vapor can be defined and demarked by a temperature-dependent surface tension. The vapor bubble continues to expand and will reach a radius of critical size, as defined by Eq. (1), if the initial neutron-nucleus interaction has been sufficiently energetic for the given degree of superheat. Apfel, Chu, and Mengel have found that only a relatively small percent of the deposited energy remains inside the bubble during this process, which is consistent with the low values of η calculated from our data and that of Greenspan and Tschiegg.

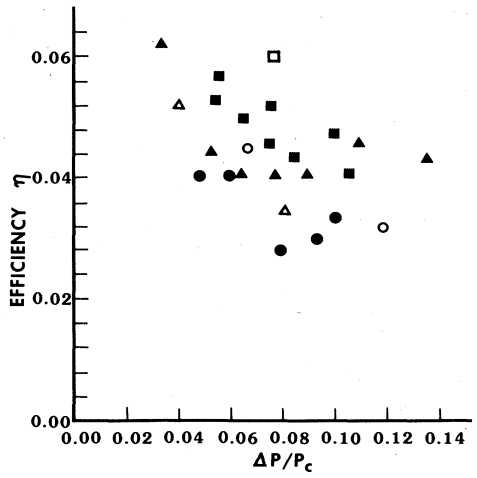

FIG. 3. The efficiency, η, of bubble production as defined in Eq. (4) is plotted against the maximum pressure difference across the bubble interface (normalized to the critical pressure) for two cases: (a) neutron-induced acoustic cavitation in *n* pentane, (●); Freon-11, (▲); and Freon-113, (■); and (b) neutron-induced nucleation of superheat Freon-114, (△); isobutane, (○); and Freon-12, (□).

It is interesting to note from the nature of the (dE/dx) curve (Figs. 1 and 2) that one cannot deposit more than a certain maximum amount of energy per unit length by increasing the neutron energy; this maximum corresponds to the maximum value of (dE/dx) for a given ion in a given liquid. In other words, by using the maximum values of (dE/dx) and an approximate value of η, one can estimate that temperature below which no neutrons, however energetic, can induce vapor nucleation. We calculated this "inhibition temperature" for two different liquids (Freon-114 and isobutane) assuming that an elastic, head-on collision by neutrons is the mechanism for energy transfer. For both liquids we assumed an η value based on the measured result for the threshold at the lower temperature. For isobutane, carbon is the relevant ion that produces the maximum stopping power; for Freon-114, the relevant ion is chlorine. These inhibition temperatures are also presented in Tables I and II.

In another set of experiments we observed that the thermal-neutron sensitivity of Freon-12 is lost at temperatures below about 10 °C. It has been proposed[19] that the sensitivity of Freon-12 to thermal neutrons is due to the following nuclear (n,p) reaction

$$^{35}_{17}\mathrm{Cl} + ^{1}_{0}n = ^{35}_{16}\mathrm{S} + ^{1}_{1}\mathrm{H} \tag{5}$$

(and not by the usual elastic head-on collision); the proton deposits 598 keV as it travels through the liquid. The sulfur ion deposits its entire 17 keV in a range that is typically less than a critical radius, R_c. The cross section for reaction in Eq. (5) is about 0.4 b. [We also considered the $Cl^{35}(n,\alpha)P^{32}$ reaction until we realized that the cross section for this reaction was four orders of magnitude less than the $Cl^{35}(n,p)S^{35}$ reaction.]

The energy required to form a vapor bubble for Freon-12 at 10 °C using Eq. (3) is found to be 1.2 keV. The energy deposited in Freon-12 by the relevant ions formed by thermal neutrons through an elastic head-on collision is at least three orders of magnitude less than 1.2 keV and, therefore, the elastic head-on collision cannot be the mechanism of energy deposition for nucleation. On the other hand, at 10 °C the proton produced by the above nuclear reaction (with energy 598 keV) and the sulfur ion (with energy 17 keV) deposit approximately 19.2 keV in a critical diameter about the point of the inelastic collision. This corresponds to a value of η of about 6%, which is in the upper part of the range of η values given in Fig. 3. These calculations confirm that the above nuclear reaction

appears to be the likely mechanism of energy deposition and hence the cause of the thermal sensitivity of Freon-12.

SUMMARY AND CONCLUSION

We have shown that with knowledge of the stopping power and the critical radius associated with a superheated liquid, we can make reasonable estimates of the threshold energy at which neutrons will nucleate the vapor-phase transformation. The results have relevance to the design of a neutron detector based on superheated drops in gel.[20] The present study also indicates that a small fraction of the deposited energy is used up for vapor-bubble nucleation and implies that a more correct theory including all dynamic factors involved in the process must be developed before a more fully predictive theory is possible.

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