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THE SUPERHEATED DROP DETECTOR

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The Superheated Drop Detector (SDD) is a new tool for radiation spectrometry, area monitoring, and dosimetry that may find use in nuclear science, safety, and medicine. It is based on the principle of the bubble chamber, but unlike the bubble chamber, offers continuous radiation sensitivity, portability and adaptability, direct reading capability, and low cost. Moderately superheated drop detectors are insensitive to gamma rays and X-rays for energies less than 6 MeV, but sensitive to fast neutrons and have an energy threshold that can be adjusted by varying temperature, pressure, or drop composition.

1. Introduction

It is well known that liquids can be superheated, but this metastable state is generally observed to be fragile and short-lived owing to the abundance of microscopic gas pockets and bubbles (heterogeneous nucleation sites) at liquid-solid interfaces either on particles or container surfaces. Only *one* of these sites need be present to act as the seed for the growth of a vapor bubble and, hence, the termination of the superheated state. Even in the absence of these sites, the natural radiation background will be responsible for the termination of the superheated state in a relatively short time if the sensitive volume is large.

It is, therefore, not surprising that even the "clean" bubble chambers, first developed by D. Glaser, seldom survived significant superheating for more than a few minutes[1]). In practical bubble chambers the liquid is first superheated by briefly dropping the pressure below the liquid's vapor pressure; the nuclear event is then recorded by the trail of bubbles nucleated by elementary particles; and then the chamber is rendered stable again by re-pressurization. Typical repetition rates for bubble chambers are less than five pulses per second[2]).

Although occasionally used in neutron spectrometry[3]), little serious thought has been given to the idea of using the bubble chamber for dosimetry, area monitoring, and practical spectrometry. For these applications it is often desirable to have a much simpler and portable, integrating detector.

The present idea for such a detector grew out of the ideas of Lieberman[4]), Finch[5]), Hahn and Peacock[6]), Greenspan and Tschiegg[7]), Bertolotti et al.[8]) and West and Howlett[9]), who considered radiation-induced acoustic cavitation, and that of Hahn and Spadavecchia[10]), who used negative pressure to sensitize liquids. The major limitation of these techniques is the difficulty of generating and maintaining large tensile stresses in liquids. Rarely have cavitation measurements given the tensile strength of the pure liquid[11,12]). Furthermore, cavitation bubble collapse in a bulk sample often produces large numbers of bubble "seeds" which weaken the liquid to future tensile stresses.

Experiments with superheated drops rising in a heated "host" liquid, on the other hand, have lead to repeatable and reliable results for the limit of superheat of liquids[13-15]). This success has been attributed to the subdivision of the sample into drops (which assures that some will be free of nucleating impurities and that radiation triggering one will not trigger all) and to the suspension of the sample in a "perfectly smooth" container – another liquid.

Since rising drops are not particularly convenient to work with in nucleation experiments, researchers have tried a number of techniques for immobilizing drops, from sandwiching samples between two different and mutually immiscible liquids[16,17]), to levitating drops by flow-drag[18]) or acoustical means[12]). Dispersing drops in a gel-like medium has proven a simple and economical solution. The drops are simply introduced at a temperature below the liquid's boiling point, and then the temperature is increased to the desired degree of superheat; alternatively the drops can be introduced at a pressure greater than the liquid's vapor pressure, and the desired degree of superheat achieved by reducing pressure. The details of the procedure for assuring such long-lived, superheated drops is detailed elsewhere[19]).

Superheated drops in a gel-like host are continuously sensitive, miniature, bubble chambers. By subdividing the sample into drops it has been assured that one nucleation event does not consume the sample, and, therefore, the re-pressurization procedure for traditional bubble chambers is avoided. Such a detector needs no power source, because the drops themselves represent stored mechanical energy which is released when triggered by radiation. Furthermore, the total amount of vapor evolved from the radiation-induced nucleation of drops can serve as a convenient integrated measure of the total exposure of the detector to a particular type of radiation above a "threshold" energy.

2. Theory of operation

The theory of radiation-induced nucleation of superheated drops involves the nuclear physicist (if the interaction of an uncharged particle with a nucleus is being considered), the atomic physicist for the interaction of ions with matter, and the fluids physicist for describing the dynamic process resulting in a macroscopic vapor bubble. Since this "event" includes different physical phenomena occurring over a length scale covering twelve orders of magnitude, it should not be surprising that no existing theory has been capable of making accurate absolute estimates of, for example, the threshold energy of neutrons required to nucleate bubbles in a given liquid superheated to a known degree.

The problem has not only been with theory, but also with the scarcity of experiments using a variety of liquids which measure nucleation conditions over a wide range of nuclear, fluid, and thermal parameters. This data void may be filled, in part, by experiments using calibrated superheated drop detectors (SDDs).

A model for neutron-induced vaporization of superheated drops is proposed below[20]).

Neutrons of energy E_n and flux ψ (neutrons/cm$^2\cdot$s) are incident on N superheated drops of total volume V, liquid density ρ, and molecular weight M. As long as the total number of drop vaporization events is small compared to N, the vaporization interaction rate (number/s) is given by

$$\phi = \psi V \sigma(E) \cdot N_0 \rho / M , \qquad (1)$$

where N_0 is Avogadro's number and $\sigma(E)$ is an effective cross section for interaction per molecule (typically on the order of 1 b). $\sigma(E)$ is more complicated than traditional nuclear cross sections, not only because we are dealing with molecules rather than atoms, but more importantly, because an "event" depends not only on neutron–nucleus interaction but also on whether the interaction leads to a macroscopic bubble; thus σ depends both on the neutron energy and the degree of superheat of the liquid.

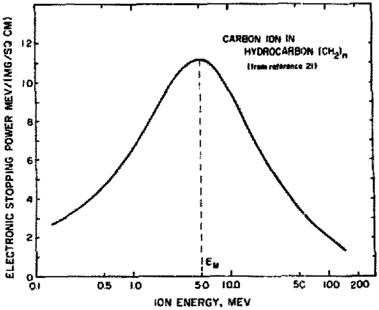

Fig. 1. Typical shape of stopping power curve, as exemplified by this curve for carbon ions in hydrocarbons of the form $(CH_2)_n$. (Data from ref. 21.) The greatest stopping power occurs when ions slow due to electron collisions to an energy E_M. Near this energy charge capture commences. If an ion's initial energy is less than E_M, its local energy deposition is greatest at the beginning of its track.

The maximum amount of energy that the nucleus (atomic weight A, in amu) can receive from the neutron is $4AE_n/(A+1)^2$ – from a head-on elastic 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. The rate at which energy is lost by the ion is traditionally referred to as the stopping power, $-\mathrm{d}E/\mathrm{d}x$, and has the familiar shape shown in fig. 1. For a given incident ion energy, E_i, the range of the ion, before stopping, can be found from $R = \int_{E_i}^{0} (-\mathrm{d}E/\mathrm{d}x)^{-1}\,\mathrm{d}E$. Tables by Northcliffe and Schilling[21]) give $-\mathrm{d}E/\mathrm{d}x$ and R, but these may be inaccurate if the relevant initial ion energy is near or below the energy at which the stopping power has its maximum value, E_M [22]). This may be the case for neutron dosimetry.

There has been some controversy about mechanisms for vapor bubble nucleation, but it is generally agreed that Seitz's thermal spike theory[23]) is appropriate[24]). In this theory it is presumed that the atomic agitation along the path of the ion is equivalent to a hot spot which literally explodes, creating a sufficiently large vapor bubble. Initially behind the shock wave is a "fluid" at conditions of temperature and pressure far beyond the critical parameters of the liquid; but as the shock wave moves out, the region cools and finally at some position within the shock front the critical parameters are realized and an interface separating liquid and vapor can be defined. This vapor bubble will grow, but adiabatic cooling, heat conduction, viscosity, and surface forces will gradually take away its "steam". The bubble becomes large enough to nucleate the macroscopic liquid-to-vapor transformation if the vapor cavity reaches a critical radius R_c defined by

$$R_c = 2\gamma(T)/\Delta P, \qquad (2)$$

where $\gamma(T)$ is the surface tension and where $\Delta P = P_v(T)+P_g-P_0$, with P_v the pressure of the vapor in the cavity, P_g the partial pressure associated with non-condensible, dissolved gas, and P_0 the externally applied pressure. ΔP is one measure of the degree of superheat of the liquid, and defines the critical size. For larger ΔP, R_c is smaller, and the heat required for drop vaporization is less.

The heat deposited per unit length is determined from stopping power data for a given ion traversing the liquid. In the case of neutron radiation, the nucleus of an atom of the liquid with which it interacts becomes the ion. Although heavier ions receive less energy from the neutron than lighter ones, they are more efficient at depositing their energy except at energies much below E_M (see fig. 1).

Only the energy along that part of the ion's range corresponding to about twice the critical radius will contribute significantly to bubble formation, and only a fraction of that energy is effective in producing the bubble of critical size.

3. Experimental procedure and preliminary data

In this section we discuss measurements of bubble nucleation in three liquids: CCl_2F_2 and $C_2Cl_2F_4$ (often referred to as refrigerants 12 and 114, respectively) and cis-2-butene, C_4H_8. Neutrons of known energy were produced via a d–d reaction using a 1 MeV Van de Graaff accelerator (Sloane Physics Laboratory, Yale Univ.). The SDD samples were in a vial which was, in turn, encased in a flexible plastic bag so that the only gas in contact with the gel was that of the drop's vapor. (A pinhole in the plastic acted as a pressure release.) The samples were in a temperature and pressure controlled lucite chamber (see fig. 2). The measurement procedure consisted of irradiating the sample with neutrons while reducing the pressure P_0 in steps of about 0.05 atm. every 2 min until the 1–3 mm drops were observed (via CCTV) to vaporize. Results were repeatable to about 0.05–0.1 atm. The temperature T was held constant within 0.25 °C.

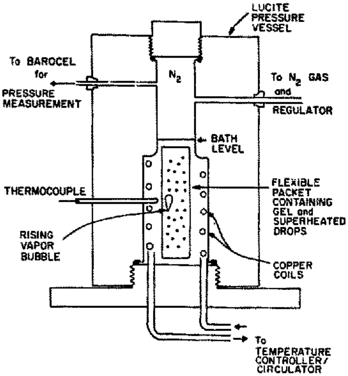

Fig. 2. Schematic of apparatus used to measure the pressure threshold at which a given energy neutron will nucleate bubble formation in a superheated liquid at a fixed temperature.

Table 1 displays some preliminary data from these experiments. Column 4 gives the degree of superheat as measured by the difference of the vapor pressure and the measured threshold pressure, P_0. The critical radius calculated according to eq. (2) is in column 6, and the reversible work to form a cavity of this size is calculated in column 7. [This is just the thermodynamic work to form the surface, $4\pi\gamma R_c^2$, and to expand the volume, $-\frac{1}{3}\Delta P 4\pi R_c^3$, with R_c given by eq. (2).] Since the heat deposited per unit length is an important parameter in the thermal spike model, the maximum possible energy deposited in a critical diameter ($2R_c$) has been calculated for all possible ions of the liquid; it was found that for all three liquids, the carbon ion deposited the most energy E_D in the critical diameter (column 9). (The initial en-

TABLE 1

Preliminary data.

Liquid	Temp. (°C)	Threshold pressure P_0 (atm)	$\Delta P = P_V - P_0$ (atm)	Surface tension (dyn/cm)	Critical radius, R_c (μm)	Reversible work, $W = \frac{16\pi\gamma^3}{3\Delta P^2}$ (keV)	Initial ion energy, E_i (MeV)	Deposited energy, E_D (keV)	W/E_D (%)
1	2	3	4	5	6	7	8	9	10
CCl_2F_2 refrigerant 12	34.3	7.40	0.81	8.0	0.20	0.82	1.18	211	0.39
$C_2Cl_2F_4$ refrigerant 114	34.3	1.82	1.09	11.1	0.21	1.20	1.18	269	0.45
C_4H_8 cis-2-butene	34.2	1.32	1.49	13.5	0.18	1.16	1.18	152	0.76

ergy of the carbon ion, received from an elastic, head-on collision with a 4.14 MeV neutron is shown in column 8). Column 10 gives the reversible work to form a critical bubble as a percent of the deposited energy, and is therefore a measure of the relative efficiency of the heat deposition process in bubble formation.

There are two interesting comparisons to be made. The two refrigerants have nearly the same critical radius (even though the measured threshold pressure for one is four times that of the other). This implies that one might calculate the threshold pressure of one if one knew the threshold pressure of the other; evidently, the dynamics of the process will be similar for the same family of liquids. The second comparison comes between refrigerant 114 and cis-2-butene. These liquids have nearly the same boiling points and vapor pressure curves. Their critical radii are a little different; the energy deposited in the critical diameter of cis-2-butene is only 57% that of refrigerant 114, but the degree of superheat as measured by ΔP is 37% greater for cis-2-butene. Clearly in this case the similarity of critical radii points to the greater superheat required for cis-2-butene, but dynamic factors will have to be brought in for quantitative comparisons.

4. SSD sensitivity to X-rays, gamma-rays, and charged particles

From the above, it is clear that the efficiency of energy deposition is a crucial factor in triggering bubble formation. It is not surprising, then, to find that for moderately superheated liquids (ΔP less than 5 atm), superheated drop detectors are insensitive to gamma rays and *X*-rays below 6 MeV. In this energy range gammas interact with electrons and are inefficient in depositing their energy locally. Above 6 MeV, when the gamma energy is comparable to the binding energy per nucleon, the probability of photonuclear interactions increases, and heavy ions with sufficient energy are produced.

SDDs will also be sensitive to charged particles if they are sufficiently energetic to traverse the host gel and container materials. Further work must be done in this area.

5. Practical features of SDDs

Since superheated drops represent stored mechanical energy, and since the probability of interaction is proportional to the sensitive volume, the volume of vapor evolved from radiation-induced interactions is an integrated measure of exposure to above-threshold radiation.

This vapor can be measured directly in a number of ways, from the size of a collected vapor bubble to the height that a hydrometer-like device floats above a reference point (see fig. 3). These and other possible embodiments are discussed in ref. 19.

If information about the energy spectrum of the radiation is desired, the simplest approach is to use several SDDs, each with a different composition (and thus different known energy threshold). Measurement of the amounts of vapor produced in a given time interval in the separate detectors can be used to determine the amount of radiation in the energy intervals defined by the energy thresholds of the detectors. (SDDs can be stacked

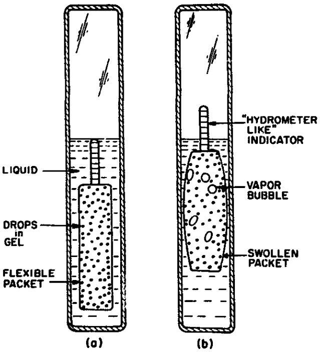

Fig. 3. Hydrometer-type embodiment of SDD, (a) before radiation; (b) during radiation drops vaporize, and the flexible packet expands and rises (due to buoyancy) in the surrounding liquid. The distance the indicator rises above the liquid level is one measure of radiation exposure. (Taken from patent application, ref. 19.)

or sandwiched like detectors using nuclear photographic emulsion.)

This kind of spectrometry can also be performed with several detectors of the same composition if the energy threshold of these detectors is adjusted to different values by varying the temperature and/or pressure. If the pressure is varied, one must take care not to increase the dissolved gas concentration in the gel, because this will decrease the energy threshold [see eq. (1)] by increasing the effective degree of superheat.

6. Areas of possible application

Because of the simplicity of composition and the lack of required electronics, superheated drop detectors should be able to be designed in a number of different formats for different applications. As with the bubble chamber, sensitivity goes as the sensitive volume, and, therefore, compact drop detectors are rather inefficient. Nevertheless, long term monitors and dosimeters (e.g. 2–4 week) can be conveniently small (e.g. shirt pocket and possibly even "badge" size). Short term units requiring high interaction rates will be correspondingly larger. For area monitoring, large "sheets" of detector composition could be hung in critical places. This sheet could be compartmentalized, each compartment serving as a little detector. Such a detector could be monitored at a distance if a color dye were incorporated into the sensitive material.

For potentially hazardous work, the lack of power supply, the ease of handling, direct reading capability, and the simple and reliable nature of such a device provide incentives for its adoption.

SSDs also show promise as a research tool for studying the radiation–matter interaction. For instance, if the thermal spike model turns out to be a good predictor of thresholds for radiation-induced vaporization, then one should be able to infer information on the stopping power of the sensitive material from the results of threshold measurements. Isotopes and isomers will be particularly good candidates for probing the radiation-matter interaction.

7. Potential disadvantages

Although this new detector shows promise because of the features discussed above, there are some problems and limitations that may discourage its practical use in some areas.

The advantage of a temperature dependent energy threshold for use in spectrometry carries the disadvantage to the user who must use a SDD in different thermal environments.

The advantage of ease of use and portability may be offset, in part, by the fact that SDDs are not reusable; one cannot condense the vapor back into drops if it separates from the gel. This may not be too bad, because the cost of the materials in a disposable packet of a small detector is on the order of pennies.

Since the detector is continuously sensitive if superheated, storage in a freezer or under pressure is appropriate before or in between use so that the ambient radiation background does not trigger drops. Not only does radiation trigger drops; any microscopic gas pocket that comes into contact with a drop will nucleate the vapor phase. Outgassed gel should be used in the formulation, a prepressurization process for eliminating potential gaseous nucleation sites should supplement the outgassing[25], and the composition should be sealed to gases other than the vapors of the drop and gel materials. In addition, one must make sure that the vaporization of one drop does not trigger another either by mechanical disturbance (the gel is a good shock absorber) or by leaving a potential nucleation site around. Because of the newness of the device, long term changes in composition or sensitivity have not been assessed.

8. Concluding remarks

Most notably not reported here are comprehensive measurements of cross sections of interaction [eq. (1)]. These may be forthcoming with the assistance of the neutron calibration facility of the National Bureau of Standards. Though much of this work is in its infancy, it was considered appropriate to report briefly this initial progress in the hope of stimulating suggestions for the SDD's future development and practical application to the needs of nuclear science.

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