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LET dependence of bubbles evaporation pulses in superheated emulsion detectors

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ABSTRACT

Superheated emulsion detectors are suspensions of metastable liquid droplets in a compliant inert medium. Upon interaction with ionizing radiation, the droplets evaporate, generating visible bubbles. Bubble expansion associated with the boiling of the droplets is accompanied by pressure pulses in both the sonic and ultrasonic frequency range. In this work, we analyzed the signal generated by bubble evaporation in the frequency and time domain. We used octafluoropropane (R-218) based emulsions, sensitive to both photons and neutrons. The frequency content of the detected pulses appears to extend well into the hundreds of kHz, beyond the range used in commercial devices to count bubbles as they are formed (typically 1–10 kHz). Kilohertz components characterize the early part of the waveforms, potentially containing information about the energetics of the explosive bubble initial growth phase. The power spectral density of the acoustic signal produced by neutron-induced evaporation shows a characteristic frequency pattern in the 200–400 kHz range, which is not observed when bubbles evaporate upon gamma ray-induced irradiation. For practical applications, detection of ultrasonic pulses associated with the boiling of the superheated drops can be exploited as a fast readout method, negligibly affected by mechanical ambient noise.

1. Introduction

Superheated emulsions (SE) consist of droplets of a halocarbon, suspended in an inert, compliant gel matrix. At room temperature ($\approx 20\,^{\circ}$C) and inside a defect-free matrix, the drops are kept in a metastable superheated liquid state, i.e. above the boiling point and below the critical point of the halocarbon (Table 1). SE were first used for radiation detection purposes in the late 1970s by Apfel [1], who observed that ionizing radiation triggers metastable droplet transition from liquid to vapor phase. The bubble chamber also exploits liquid's momentarily superheated state to track an elementary particle's path by the bubbles it nucleates as it traverses the liquid [2,3]. However, unlike bubble chambers, SE are continuously sensitive to radiation, since liquid droplets are permanently kept in the superheated state. Nucleation of vapor bubbles requires a minimum amount of energy to be deposited within a critical length. SE detectors thus show a threshold radiation response, which is primarily related to the linear energy transfer (LET) dE/dx of the interacting charged particles [4].

Bubble nucleation is a cavitation process associated with high internal pressures, exceeding 10 MPa, and high temperatures of several thousand degrees Kelvin, and leads to observable phenomena such as shock waves and thermal effects (fusing) [5]. The first phase of the evaporation involves the generation of a shock wave resulting from the heating of a small region to temperatures and pressures exceeding their critical values. A direct method for real-time counting of the number of evaporated bubbles relies on the acoustic detection of oscillating pressure waves accompanying bubble expansion. These oscillating pressure pulses show a principal harmonic of the order of a few kilohertz (e.g. 5 kHz for R12 [6] and 2.8 kHz for R500 [7] based emulsions), which can be easily recorded with commercial low-cost piezoelectric sensors. For this reason, acoustic counting was among the first readout approaches of superheated emulsion detectors. Currently, sensors centered at 5 kHz are used in a double piezoelectric transducer configuration, where true vaporization events can be distinguished from mechanical noise both by anticoincidence and by pulse shape analysis [8]. This configuration allows operation with ambient noise levels above 100 dB. However, recent studies revealed a significant component of the frequency spectrum of the pressure pulses above 5 kHz for C-318 [9], R-115 [10] and R610 [11,12]. Bubbles generated after neutron interactions can also be discriminated from those produced either by photons [13] or alpha particles [10,14], since pressure pulses accompanying bubble evaporations carry

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Table 1
Properties of superheated liquids typically used for radiation detection. Reduced superheat, critical radius and E_c energy deposited by carbon recoils in the critical diameter are calculated for a temperature of 22 °C [18].

Halocarbon refrigerant code	Chemical formula	Boiling point (°C)	Critical point (°C)	Reduced superheat s	Critical radius R_c (μm)	E_c[a] (En=0.1 & 10 MeV) (keV)	Radiation sensitivity at room temperature
R-114	$C_2Cl_2F_4$	3.65	145.70	0.13	0.243	72.10 371.06	Heavy ions and fast neutrons
C-318	C_4F_8	−6.99	115.22	0.24	0.095	37.92 160.73	Heavy ions and fast neutrons
R-610	C_4F_{10}	−1.7	113.2	0.21	0.077	29.59 129.84	Heavy ions and fast neutrons
R-115	C_2ClF_5	−39.17	79.95	0.51	0.014	3.12 18.34	Light ions, photons and neutrons
R-218	C_3F_8	−36.65	71.95	0.54	0.012	4.03 17.86	Light ions, photons and neutrons

[a] $E_c = dE/dx\, 2R_c$.

different spectral and amplitude features in the two cases. In fact, the analysis of high frequency components of pressure pulses generated by bubble cavitation may allow the characterization of the bubble evaporation dynamics. For practical applications, ultrasonic detection as a readout method has two main advantages: a lower measurement dead-time, reduced by a factor of about 100 compared to the low-frequency counting approach; and insensitivity to mechanical external noise. Although the pressure wave attenuation in fluids, according to the classical theory, is proportional to f^2, pressure waves in the kilohertz frequency range can be effectively detected by placing the sensor right in contact with the detector gel matrix (speed of sound in the medium 1920 m s^{-1}). The purpose of this study is to confirm previously published works stating the presence of high frequencies and extend their analysis to octafluoropropane (R-218) in the 200–800 kHz range. By the extraction of several features of evaporation pulses, we also aim to discriminate them according to the energy loss of the interacting charged secondaries (i.e. LET of the primary radiation). Radiation is typically classified as either high or low LET, based on the amount of energy it transfers per unit path length it travels. Relatively heavy recoil ions, with a mass over 1700 times that of an electron, are generated by neutrons via scattering or (n, x) reactions and produce a dense ionization path. They can be thus classified as high LET radiation. On the other hand, highly energetic electrons are produced by photons via photoelectric effect, Compton scattering and pair production are considered low LET radiation, as their mass is smaller than recoil ions' mass and results in a greater distance between collisions and thus a lower linear energy transfer. This classification holds in this context, where detectors were irradiated with neutrons (^{241}AmBe, 11 MeV neutron maximum energy) and photons (^{22}Na, yielding 1.275 MeV and 511 keV photons, and ^{137}Cs, yielding 662 keV photons). The stopping power of both recoil ions and electrons exceeds the onset energy loss per unit length for R-218 droplets evaporation at room temperature, of order ≈ 39 keV/μm at 22 °C. However, the different amounts of energy density, the spatial location and time scale of ionization and excitation interactions for recoil and electron tracks are expected to affect the nucleation dynamics and thus the frequency content of evaporation acoustic pulses.

2. Materials and methods

Seitz originally suggested that ions deposit their energy via a "thermal spike" [3] and initiate the phase transition, generating trails of vapor embryos of sub-microscopic diameter. If the energy released within the liquid droplet is enough so that mechanical equilibrium between surface tension and pressure difference through the walls of the cavity is reached, the vapor embryo reaches a "critical" radius [3] R_c and grows to observable size through the evaporation of the available superheated liquid. Different emulsions can be produced to be selectively sensitive to different radiation qualities. A direct comparison of the radiation response of different halocarbon emulsions is possible through the reduced superheat factor s (Eq. 1). It represents the normalized operating point of a liquid within the temperature range where its superheated state is allowed [15], at a given pressure.

$$S = (T - T_b)/(T_c - T_b) \tag{1}$$

In Eq. (1), T_b and T_c are the boiling and critical temperatures, respectively. For radiation detection purposes, the reduced superheat of halocarbons ranges from 0.1 to 0.6, with a virtual upper limit of 0.65 where thermal agitation makes superheated liquids intrinsically unstable and homogeneous nucleation occurs without external irradiation. The standard conditions for pressure apply for these reduced superheat values and their subsequent results [16]. Reduced superheat can be used to predict SE energy response: the higher the degree of superheat, the lower the energy required for the radiation-induced nucleation. For low values of reduced superheat, only high-energy ions, such as the recoils generated by fast neutrons, are able to trigger drop evaporation, with a linear energy transfer of hundreds of keV per micron. For example, emulsions of R-114 (s=0.13 at 22 °C) and C-318 (s=0.23 at 22 °C) at room temperature are sensitive to fast neutrons and insensitive to photons [17]. Conversely, photon sensitization arises for a reduced superheat above 0.45, which can be achieved either in R-114 and C-318 by increasing the operational temperature, or at room temperature using liquids which are highly superheated, such as R-115 (s=0.51 at 22 °C) and R-218 (s=0.54 at 22 °C). C-318 and R-114 become sensitive to photons at about 70 °C. R-115 and R-218, instead, show photon sensitization already at 16 °C and 13 °C respectively, since they require a track-averaged vaporization energy of tens of kiloelectronvolts per micron, which can be delivered also by light ions and secondary electrons produced by photon interactions. Table 1 shows the main thermodynamic and detection properties of the abovementioned superheated liquids, together with the energy deposited in the critical diameter (E_c) by neutron recoil ions, calculated at 22 °C. The table reports E_c for carbon ions receiving the maximum energy after head-on collision with 0.1 and 10 MeV monoenergetic neutrons. Carbon ion has been selected since it deposits the most energy in a critical diameter, for the listed liquids at 22 °C, with respect to fluorine and chlorine ions. In our experiment, electrons with an average energy of about 300 keV are produced within the detector mainly via Compton scattering with ^{22}Na and ^{137}Cs photons. For the two compounds R-115 and R-128, the energy deposited in the critical diameter by these electrons at the end of their path (≈ 1.1 keV [6])