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Bubble Radiation Detection: Current and Future Capability

Anthony J. Peurrung
Richard A. Craig

November 1999

Prepared for the U.S. Department of Energy
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Summary

Despite a number of noteworthy achievements in other fields, superheated droplet detectors (SDDs) and bubble chambers (BCs) have not been used for nuclear nonproliferation and arms control. This report examines these two radiation-detection technologies in detail and answers the question of how they can be or should be "adapted" for use in national security applications.

These technologies involve closely related approaches to radiation detection in which an energetic charged particle deposits sufficient energy to initiate the process of bubble nucleation in a superheated fluid. These detectors offer complete gamma-ray insensitivity when used to detect neutrons. They also provide controllable neutron-energy thresholds and excellent position resolution. SDDs are extraordinarily simple and inexpensive. BCs offer the promise of very high efficiency (~75%). A notable drawback for both technologies is temperature sensitivity. As a result of this problem, the temperature must be controlled whenever high accuracy is required, or harsh environmental conditions are encountered.

The primary findings of this work are listed and briefly summarized below:

- SDDs are ready to function as electronics-free neutron detectors on demand for arms-control applications. The elimination of electronics at the weapon's location greatly eases the negotiability of radiation-detection technologies in general.
- As a result of their high efficiency and sharp energy threshold, current BCs are almost ready for use in the development of a next-generation active assay system. Development of an instrument based on appropriately safe materials is warranted.
- Both kinds of bubble detectors are ready for use whenever very high gamma-ray fields must be confronted. Spent fuel MPC&A is a good example where this need presents itself.
- Both kinds of bubble detectors have the potential to function as low-cost replacements for conventional neutron detectors such as ^{3}He tubes. For SDDs, this requires finding some way to get boron into the detector. For BCs, this requires finding operating conditions permitting a high duty cycle.

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1.0 Introduction

The advent of the first bubble chambers (BCs) in the 1950s brought tremendous advantages to physicists seeking to detect and understand the nature of the exotic particles being discovered at the time. Recently, the advent of superheated droplet detectors (SDDs) based upon a closely related technology has provided a new class of dosimeters that is finding use for a variety of applications (Ing et al. 1997). This report seeks to determine whether these technologies can be used for applications in nuclear nonproliferation and arms control. Based upon a detailed examination of the physics and current capabilities of these instruments, we determine what applications they might address in their current forms and what applications they might address assuming the execution of research and development aimed at adapting them to national security needs.

Nearly every radiation-detection technology involves some physical method for "exaggerating" the effect of a radiation interaction to render it more readily observable. BCs and SDDs accomplish this using a superheated fluid in which a bubble nucleates at the site where decelerating charged particles deposit energy. The two types of bubble detectors (BCs and SDDs) differ in the way in which the superheated fluid is prevented from boiling spontaneously. BCs achieve this by detecting radiation only in the moments after the fluid becomes superheated as a result of a rapid pressure change. This report takes as a basic property of BCs, namely, that they must be cycled between superheated and non-superheated states. SDDs prevent spontaneous boiling by using only droplets of superheated fluid that are so small that surface tension prevents bubble nucleation.

A motivation for this work is that the common properties of all bubble detectors make them potentially well suited for a much wider variety of radiation-detection applications than those for which they are now used. Because the process of bubble nucleation is highly sensitive to the amount of energy deposited per unit track length, detectors are inherently capable of highly effective particle discrimination. The rate of energy deposition (dE/dx) is a strong function of the charge-to-mass ratio of the decelerating charged particle. Thus, bubble detectors can be made to detect protons but not electrons (neutrons but not gamma rays), electrons but not muons, alpha particles but not protons, etc. Because dE/dx is an increasing function of energy for the recoil nuclei, bubble detectors can easily be made sensitive only to neutrons above a certain energy threshold. When used for neutron detection, bubble detectors are particularly insensitive to gamma rays, with the ratio of sensitivities for the two types of radiations exceeding 10^{12}. Another desirable property of all BCs is their inherent position resolution. Because bubbles have an acoustic and optical signature that can be easily recognized and located, they hold great potential for radiation imaging.

The remaining discussion and analysis of BCs and SDDs is best separated and is therefore contained in Sections 2.0 and 3.0, below. Section 4.0 concludes by describing the applications for bubble detectors in the area of nonproliferation and national security, along with an outline of the research and development necessary to enable these applications to succeed.

2.0 Superheated Droplet Detectors

A superheated droplet detector consists of a suspension of superheated droplets in a polymer matrix.[1] Although a variety of materials have been used for the superheated droplets, the various types of Freon are a common choice because of their appropriate physical and chemical properties. Literature values for the droplet volume fraction vary from 0.01% to 10%. The droplet diameters are between 10 and 100 microns, with 25 microns a typical value. Index-of-refraction matching between the polymer matrix and the droplets often ensures that the medium remains transparent rather than turning white like milk or snow as a result of internal refraction. While the detector sizes are typically 4 to 8 cm^3 for the common application of dosimetry, much larger detectors with volumes up to 250 cm^3 have recently been produced and tested. Two different companies sell a variety of detectors with different sensitivities, energy thresholds, sizes, and approaches to bubble counting.[2]

An understanding of the bubble nucleation process is helpful for understanding the radiation response of these detectors. While the original theory of radiation-induced bubble nucleation developed for BCs (Section 3.0) remains essentially valid (Seitz 1958) several refinements and applications to the problem of droplet detectors have been added by later authors (Apfel et al. 1985; Harper and Rich 1993; Harper and Nelson 1993). For a fluid in which the degree of superheating is not too severe, surface-tension forces create an activation energy that prevents immediate bubble formation. The energy released by boiling fluid varies as the cube of the bubble radius. The energy required to expand the surface of the bubble varies as the square of the bubble radius. Consequently, surface tension will re-compress all sufficiently small bubbles. Bubbles beyond a critical size will, however, continue to expand as the energy released by boiling fluid overwhelms the forces of surface tension. Notably, this critical radius is roughly 100 nm, which is far smaller than the range of many charged particles. Thus, the criterion for bubble formation can be stated as follows: a radiation interaction must deposit enough energy within a span of roughly 100 nm to form a bubble of roughly the same size. The required energy depends upon the temperature, pressure, and choice of materials, but is only 16 keV for Freon-12 at 20°C and one atmosphere (Harper and Nelson 1993).

The detector designer has considerable freedom in how to react to the process of bubble formation. For a sufficiently inexpensive detector, the bubbles can be counted by eye and the detector thrown away. (A count of roughly 50 bubbles is optimal for this.) Bubbles can also be counted acoustically via the "popping" sound emitted when they form. An optical transmission measurement also provides a simple, automated measurement of the number of bubbles that have formed. If desired, an application of pressure can be used to recompress the bubbles and enable the detectors' re-use. When operated properly in this way, years of operation can be obtained without significant changes in the detector sensitivity.

There are a number of different interaction mechanisms that can produce bubbles within a particular detector. Table 1 shows the typical range, energy, and maximum dE/dx for a variety of radiation types in hydrogenous materials with densities near 1 g/cm^3. As expected, the maximum energy deposited in 100 nm depends strongly on the charge-to-mass ratio of the particle and somewhat on the particle's energy. (There is a maximum in dE/dx as a function of energy; this maximum occurs at roughly 30 MeV for chlorine ions and at roughly 100 keV for protons.)

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[1] In essence, each droplet is a miniature bubble chamber; the plastic matrix restricts the ultimate growth of the bubble.

[2] Those companies are Apfel Enterprises, 25 Science Park, Box 4, New Haven, CT 06511, USA and Bubble Technology Industries, Inc., Highway 17 West, PO Box 100, Chalk River, ON K0J 1J0, Canada.

Table 1 Typical Energy, Range, and Maximum 100-nm Energy Loss for Various Types of Radiation

	Energy	Range	Max 100-nm Energy Deposition
Electron	200 keV	0.3 mm	5 keV
Proton	500 keV	0.01 mm	10 keV
Alpha	5 MeV	0.02 mm	15 keV
Chlorine	100 keV	200 nm	50 keV
Fission Product	100 MeV	0.01 mm	1 MeV

Based upon the data in this table, an understanding of droplet-detector response to a variety of radiations can be formulated.

- Electrons, Gamma Rays: It is relatively hard to adjust droplet detectors for gamma-ray sensitivity because the bubble-forming energy, ~5 keV, is so low. While this has been done, the resulting products are highly temperature sensitive since the droplets must be especially close to the point where they will spontaneously boil. A gamma-ray droplet detector is commercially sold, but is recommended only for "educational use."

- Protons: While it is certainly possible to arrange conditions so that a droplet detector responds to recoil protons, this is not generally the case for several possible reasons. First, such a detector would have a relatively high temperature sensitivity since relatively little bubble-nucleation energy is provided by the proton. Second, such a detector may not exhibit the extremely high gamma-ray rejection provided by other neutron-interaction mechanisms. Finally, the energy response and sensitivity of a proton-recoil–based droplet detector would not be suited for the application of dosimetry, which has motivated most of the research and development to date.

- Alphas: Because alpha radiation cannot penetrate the container, droplet detectors are not normally used to detect alpha radiation. (Pouring the suspension directly on a uranium-containing surface does, however, make an impressive demonstration.) However, internal alpha radiation does exist and may lead to bubble formation either directly or via recoil nuclei, depending upon the conditions prevailing in the droplet detector. Note that the reaction $^{6}Li(n,t)^{4}He$ is equivalent to alpha decay.

- Heavy Nuclei: Heavy nuclei are the ideal particle for bubble nucleation and, for this reason, are favored, as will be discussed below. Particle detection via heavy nuclei should provide excellent gamma-ray insensitivity and minimized temperature sensitivity. In addition, the rate of energy deposition, dE/dx, is an increasing function of energy at energies of the order of 1 MeV and below. Consequently, the use of heavy nuclei permits energy thresholds and the development of energy spectrometers. Note that when the reaction $^{10}B(n,\alpha)^{7}Li$ is used, the ^{7}Li recoil (840 keV) is the principal contributor to bubble nucleation.

Temperature sensitivity is possibly the most serious problem with the use of SDDs. Temperature changes affect both the overall sensitivity and the shape of the energy-response curve for droplet detectors. F. d'Errico et al. concluded that a dosimetry system based upon droplet detectors was "a fairly delicate system which can be operated reliably only when environmental conditions are not extreme" (d'Errico et al. 1996). Typically, neutron-sensitive droplet detectors have an operable range of roughly 25°C with substantial (factor of 4) variations in sensitivity across the full temperature range. Assuming that the temperature can be controlled to a couple of degrees, sensitivity variations should not exceed 5%, which equals the statistical accuracy of droplet detectors. Improved temperature stability can be obtained

by using "temperature compensation." This technique employs materials that expand or contract with temperature changes to alter the detector pressure and offset the effect of temperature changes.

With few exceptions, droplet detectors have found application in the area of neutron detection generally and neutron dosimetry in particular. Depending upon the materials, temperature, and pressure, the response of a typical droplet detector to neutrons as a function of energy can take the form of any of the curves in Figure 1 (Harper and Nelson 1993; d'Errico et al. 1995; d'Errico et al. 1996; Bamblevski et al. 1996; Ing et al. 1997). The response to neutrons above roughly 250 keV arises primarily from the carbon, chlorine, and fluorine nuclei produced via neutron scattering. The response to neutrons below 250 keV arises almost completely from the reaction $^{35}Cl(n,p)^{35}S$. At these energies, recoil nuclei do not have sufficient energy to nucleate bubbles. This reaction has a cross section of roughly 0.4 barns at thermal energy (0.025 eV) and correspondingly less at higher energies. While the reaction liberates 615 keV, the recoiling ^{35}S nucleus receives only 17 keV of energy. By altering the construction of the droplet detector so that bubble nucleation requires more and more energy, response curves with progressively less response at low energy can be realized. Once more than 17 keV of energy is required for bubble nucleation, the detector stops recording low-energy neutrons. As the bubble nucleation energy is further increased, the neutron energy threshold increases further. In this way, a set of droplet detectors that can be used for crude neutron energy spectrometry can be fabricated (Harper and Nelson 1993; d'Errico et al. 1995; Bamblevski et al. 1996; Ing et al. 1997). Energy thresholds between 100 keV and roughly 10 MeV are obtainable.

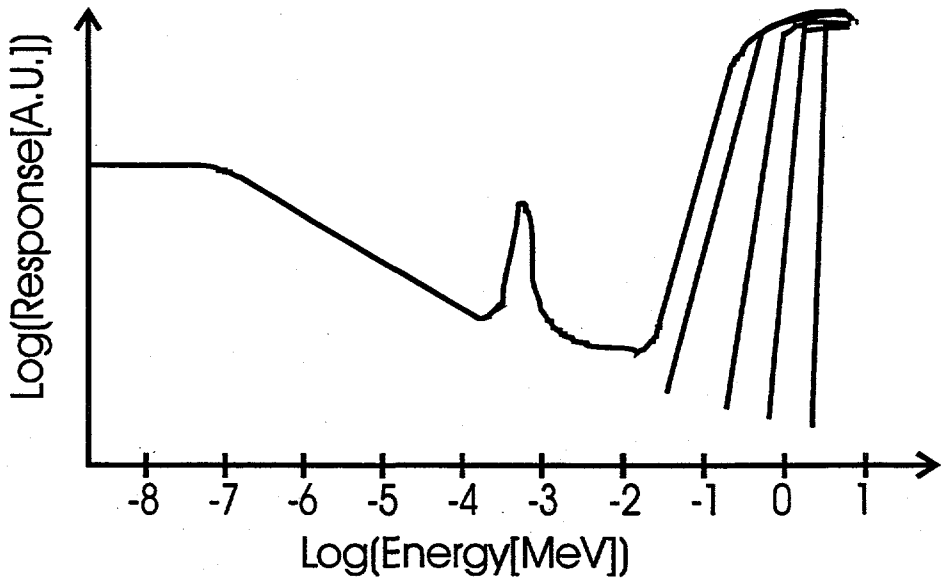

Figure 1. Possible Droplet Detector Response as a Function of Neutron Energy

The gamma-ray sensitivity of normally operated, neutron-detecting droplet detectors is essentially zero. Although the bubble-nucleation physics may be slightly different, the ratio of neutron to gamma-ray sensitivities for conventional BCs (Section 3.0) can be made to exceed 10^{12}. One experiment with droplet detectors observed no response after 400,000 R of radiation exposure from either ^{137}Cs or ^{60}Co (Schulze et al. 1992). Clearly, these detectors are able to detect neutrons in the presence of gamma-ray fluxes that would render most neutron detectors inoperable.

The neutron sensitivity of a droplet detector is a function of its size, droplet-volume fraction, and its mode of operation (temperature, pressure, droplet composition). The sensitivities in the literature are

typically between 3×10^{-5} cm^2 and 10^{-3} cm^2 for detector sizes of 4 to 8 cm^3. (These sensitivities are "flux efficiencies"—the detector count rate is obtained by multiplying by the neutron flux.) One can obtain an approximate understanding of these sensitivities via the following calculation. Assume that a detector with total volume of 4 cm^3 has a droplet-volume fraction of 0.25%. The 0.01 cm^3 of Freon in this detector has a total mass of 0.013 g and contains 0.0013 g of carbon. This quantity of carbon corresponds to 6.5×10^{20} atoms of carbon with a total cross section for fast-neutron scattering of approximately 0.0013 cm^2. Although not every recoil carbon nucleus will create a bubble, most will. Furthermore, there should be some contribution from the recoil chlorine and fluorine nuclei also present in the Freon. Thus, we conclude that the flux efficiency for this detector should be of the order of 10^{-3} cm^2, in agreement with experimental observations for the assumed detector.

The maximum flux efficiency would be obtained using a droplet detector with maximum size and droplet loading. Droplet detectors with sizes up to 250 cm^3 are commercially available. Detectors with volume fractions of 1% or greater also have been successfully constructed. (Because such detectors are not well suited for dosimetry, they remain relatively uncommon.) In principle, such a droplet detector would have a flux efficiency of 0.25 cm^2.

While the vast majority of applications for droplet detectors have been in the area of neutron dosimetry, a number of other applications have been explored. The following list describes the applications of which we are aware:

- Dosimetry
  - Medical Dosimetry: Position-sensitive droplet detectors can be inserted in medical phantoms and used to evaluate neutron radiotherapy treatments (Lim and Wang 1995).
  - Neutron Spectrometry: Droplet detectors can provide neutron spectrometric information that can be used to improve the accuracy of other neutron dosimeters (Bamblevski et al.1996; d'Errico et al. 1995).
  - Jet Aircrew Dosimetry: Droplet detectors are proposed for use as aircraft dosimeters (Tume et al. 1998).
  - Ambient Dosimetry: Highly accurate area monitors were constructed using droplet detectors (d'Errico et al. 1996).
  - Beam Contamination: Droplet detectors were used to determine the unwanted neutron dose received by patients treated via a medical electron accelerator. The excellent gamma-ray and electron rejection of droplet detectors was critical for this application (Bourgois et al. 1997; Nath et al. 1993; Ponraju, et al. 1998).
  - Gamma-ray dosimetry: Droplet detectors were evaluated and found to be unsuitable for gamma-ray dosimetry (Matiullah et al. 1992).

- Dark-Matter Detectors: Droplet detectors are well suited for dark-matter detectors since their background deep underground can be extremely low (Collar 1996; Hamel 1997).

- Spent-Fuel Monitoring: Droplet detectors are well suited for this application as a result of their excellent gamma-ray rejection (Tam et al. 1996).

3.0 Bubble Chambers

A bubble chamber is activated by suddenly reducing a liquid medium's pressure to a point where it would ordinarily boil. Radiation detection occurs during the period of time after the pressure has been reduced, but before boiling begins. While a small fraction of a second provides an ample time window for many of the previous applications for BCs, this "active" time can, in fact, be surprisingly long. Glaser (1952) was the first to demonstrate that this phenomenon could be exploited for radiation detection. He studied the boiling of diethyl ether in a smooth glass container after a reduction of pressure from 20 atmospheres to 1 atmosphere both with and without the presence of a radiation source. Note that this material was maintained at 130°C, whereas it would normally boil at a temperature of 34.6°C. In the absence of a radiation source, the liquid remained stable for up to 400 sec, with an average time of 68 sec. When a gamma-ray source was located near the superheated tube, the delay was consistently reduced to zero. The radiation was initiating boiling in the liquid.

Based on this observation, BCs were developed to observe the trajectories of fast charged particles via their interactions with the working fluid of the bubble chamber. Note that most bubbles are formed indirectly via the formation of secondary electrons (known as known-on electrons or delta rays.) Each bubble is probably the result of a single secondary electron; not all secondary electrons create bubbles. The ensemble of bubbles provides a track of the trajectory of the primary particle. The vast majority of BCs used for physics experiments operate in a condition that has been optimized for formation of bubbles by secondary electrons.

Boiling of a superheated liquid may or may not occur when a bubble is formed, depending on the size of the bubble. The forces acting on a bubble are the vapor pressure, P_v, inside the bubble, the external pressure, P, and the surface tension. The surface tension, σ, on a bubble of radius, r, provides an inwardly directed pressure given by

$$P_v = \frac{2\sigma}{r} \tag{1}$$

For a stationary bubble at equilibrium,

$$P_v - P = \frac{2\sigma}{r_e} \tag{2}$$

For bubble radii greater than r_e the vapor pressure exceeds the restoring pressures, and the bubble grows; for radii less than r_e, the restoring force exceeds the vapor pressure, and the bubble collapses. The surface energy is a function of temperature, vanishing at the critical temperature.

As a result of the above-stability condition, the process of bubble formation (nucleation) requires some initiating event or condition in order to proceed. In the absence of an external effect such as radiation interaction, bubble nucleation is the result of spontaneous statistical density fluctuations. Alternatively, bubble nucleation occurs at rough surfaces or at the location of small particles suspended in the liquid. Recently, it has been proposed (Classen et al. 1998) that free electrons produced via the ionization process repel the nearby fluid, forming a bubble that may grow or contract according to the stability criterion given above. This hypothesis for the bubble-initiation process is actually quite similar to Glaser's original model.

The most accepted model for bubble initiation is that of Seitz (1958). Seitz' model, also known as the thermal-spike model, describes the deposition of energy by secondary electrons in a small region of the superheated medium. This model predicts the amount of energy that should be required to form a stable bubble. Empirically, it is found that the required energy is significantly greater than that which Seitz predicted by addressing only static contributions to the stability criterion (Eq. 2). This discrepancy is largely resolved by carefully considering the reversible work required for bubble creation, thermal conductivity, and dynamic effects. For a typical fluid, a charged particle must deposit a critical energy of several hundred eV (E_c) in a region with a diameter twice the critical radius ($2r_c$) to nucleate a bubble. These considerations place combined restrictions on the energy of a charged particle that can initiate bubble growth. Because the energy loss per unit length is greatest for slow particles, the particle can deposit E_c in the critical volume only if its range at that energy is less than $3r_c$. (Near the end of the particle motion, the straight-line distance that the particle travels is only about 2/3 the distance it travels.)

Glaser's experiment was conducted with a "clean" container that is a container with atomistically smooth walls. Such a chamber has few or no surface sites on which to nucleate bubbles. For superheated liquids in clean chambers, the stable period can be many seconds. Maintenance of sufficiently clean conditions for large BCs operated in the mode required of physics experiments is relatively impractical. For this reason, the historical BCs used for particle physics experiments were generally operated as "dirty" chambers. These chambers rely upon the fact that the required observation period is so short that parasitic (spontaneous) boiling will not occur. These chambers are normally used with rapidly pulsed beamlines so that the period of interest is as brief as one microsecond and occurs at a known moment. Typically, the chamber operates at a temperature, T_{rad}, called the radiation temperature. A rule of thumb, which balances sensitivity with superheated liquid stability, is that T_{rad} is approximately two-thirds the difference between the atmospheric boiling temperature, T_{boil}, and the critical temperature, T_K. That is,

$$T_{rad} = T_{boil} + \frac{2\left(T_K - T_{boil}\right)}{3} \tag{3}$$

Note that the bubble chamber's working fluid may be *very* superheated under these conditions. While sensitivity to secondary electrons with energies of only a few hundred eV may be possible under these conditions, avoidance of boiling for longer than a few milliseconds with a "dirty" chamber is impossible. For propane, a typical operation temperature is roughly 60°C, which requires an overpressure of approximately 10 atmospheres to avoid boiling. For this condition, E_c is about 400 eV.

A simplified duty cycle of a convention bubble chamber consists of four periods, starting with the following elevated pressure conditions:

- the expansion period during which the pressure drops to the superheated condition. This period is as short as possible, but is limited by the sound velocity to a few ms.
- the superheated period during which measurements are made. The extent of this period is largely governed by the characteristic time that it takes for a bubble to grow from critical size ($\sim 10^{-6}$ cm) to observable size ($\sim 10^{-2}$ cm). For normal operating conditions, this time is less than 10 ms.
- the repressurization period during which the chamber pressure is re-established to the higher operating level. This period, like the expansion period, is short, and limited by the sound velocity in the liquid.
- the recovery period during which temperature homogeneity is re-established. Collapse of the bubbles results in local thermal inhomogeneities that must be removed before another cycle can begin. This period is somewhat more than an order of magnitude greater than that of the other periods so that the minimum cycle time is a few hundred ms.

The energy loss for a non-relativistic charged particle of charge Z, and velocity, v, is (Knoll 1989)

$$\frac{-dE}{dx} = \left(\frac{4\pi e^4 z^2}{m_e v^2}\right) NZ \ln\left(\frac{2m_e v^2}{I}\right) \tag{4}$$

where N and Z are the number density and atomic number of the absorber atoms, I is a characteristic ionization energy, and m_e is the mass of an electron. If we use Eq. 4 to determine dv/dx and integrate that relationship, we can get the range, R,

$$R = \int dx = M\left(\frac{4\pi e^4 z^2}{m_e}\right)^{-1} \int_{slow}^{V_0} \left(\frac{V^3}{\ln\left(\frac{2m_e v^2}{I}\right)}\right) dv \tag{5}$$

where M is the mass of the moving particle and V_0 is the initial velocity of the particle. This relationship is valid for ions until their velocity gets small enough that electron pickup (ion neutralization) becomes important. Over that range of energies, the logarithmic term in the denominator of the right-hand side of Eq. 5 is slowly varying, and R is proportional to MV_0^4 or is proportional to E_0^2/M. So for equal range,

$$\frac{E_0(\text{ion})^2}{M} = \frac{E_0(\text{electron})^2}{m_e} \tag{6}$$

This is important in light of Eq. 3 because it says that an ion with a range of $3r_c$ deposits $(M/m_e)^{1/2}$ times as much energy in that range as an electron. For the proton, this factor is roughly 40.

As noted above, bubble-chamber operation is typically optimized for observation of secondary electrons. If the operating conditions are modified such the superheat is reduced, the surface energy increases, the latent heat increases and, consequently, E_c increases. Ultimately, E_c increases to the point that no electrons can initiate growing bubbles. However, because of the conditions of Eq. 6, ions of the same energy can easily initiate growing bubble formation. This provides the opportunity for sensitive detection of ions with very high discrimination against electrons.

Unlike gamma rays, neutrons lead to the creation of energetic (keV-scale) ions through the simple process of nuclear recoil. A 1-MeV neutron that moderates in a hydrogenous medium will, on average, produce approximately 10 recoil protons with energy greater than 1 keV. If a superheated liquid is prepared such that it is insensitive to electrons, but still sensitive to protons, this neutron should initiate several growing bubbles while being insensitive to gamma rays.

If the degree of superheat of the liquid is properly controlled, the bubble chamber may be operated as a neutron spectrometer (Fisher et al. 1997). For a very small value of superheat, only neutrons above an energy threshold are able to form recoil ions with sufficient energy to nucleate bubbles. Two general approaches are possible: 1) the detector may be operated to gain insensitivity to neutrons

with energies below a selected threshold and 2) variation of the chamber's operational condition provides information on the distribution of neutron energies reaching the chamber. The probability of generating stable bubbles depends upon the incident neutron-energy distribution, the kinematical relationships governing the energy distribution of recoil ions, and the probability of generating a growing bubble as a function of ion energy. Because the distribution of recoil energies is simply determined by kinematics for a given incident neutron energy, the count rate as a function of superheat can be used to map out the neutron energy spectrum.

Bubbles, once nucleated, are relatively easy to record. In propane, roughly 1% of the critical energy goes into emission of sound waves. As with SDDs, an easily audible "popping" sound is produced by the growing bubble. In addition, bubbles larger than a few 10s of μm scatter light very efficiently. Thus, a variety of optical methods also allow for bubble detection.

4.0 Applications to Nonproliferation and National Security

4.1 Droplet Detectors

Two applications for BCs were identified that require no research and development prior to implementation. These applications are spent fuel monitoring and electronics-free neutron detection. BCs currently combine simplicity, neutron sensitivity, and gamma-ray insensitivity in a way that enables these applications under a wide variety of circumstances. The small physical size of droplet detectors also should permit them to be physically placed at the needed measurement location. Their low cost and reliability is also desirable.

The complex electronics generally necessary for radiation measurements are a major hindrance to the acceptance of radiation-detection technologies for arms-control applications. Droplet detectors offer the ability to measure gamma-ray fluxes, neutron fluxes, and neutron-energy spectra without the need for any electronics at the measurement location. This ability to perform item templating ("fingerprinting") or attribute measurements should meet with wide acceptance by alleviating concern about information security associated with the use of complex electronics.

A third identified application area for droplet detectors is low-cost neutron detection. There are clearly national security applications that would benefit from the introduction of a neutron detection technology with a cost lower than the $\$50K/m^2$ that holds today for high-efficiency detectors. Droplet detectors have the potential to lower this cost by an order of magnitude because of their relative simplicity, provided that some way can be found to increase the neutron sensitivity via the introduction of ^{10}B. (This question is discussed under the subsection on research questions and also in Appendix B.) A droplet detector containing merely 0.4 mg of ^{10}B per cm^3 of detector volume should have an efficiency at least as high as a typical ^3He-tube detector or ^6Li-fiber-containing detector. Unlike commercially available droplet detectors, such a detector would be primarily sensitive to thermalized neutrons via the reaction $^{10}B(n,\alpha)^7Li$. Neutron capture by boron would be favored because of the complete boron dispersal in the detector's hydrogenous moderating matrix. A low-cost neutron detector with an area of 1 m^2 based upon this principle need only contain the following components:

- Sealed container, ~7 to 10 cm thick
- ~1 L of Freon or Freon-like material dispersed as droplets
- 70 to 100 L of polymer matrix
- 40 g of ^{10}B, or roughly 200 g of natural boron (the cost for this much enriched boron would be ~$1000 to 2000)
- Acoustic bubble-counting mechanism (microphones, simple electronics)
- Manual reset mechanism (means to temporarily increase pressure).

4.2 Bubble Chambers

Previous research (Peurrung et al. 1998) has shown that direct fast neutron detectors retain information about a neutron's time of emission, energy, and direction that can be exploited for applications. Work in the fusion community (Fisher et al. 1997) has already demonstrated that BCs are a kind of direct fast-neutron detector and therefore can function as an enabling technology. We feel that additional applications of interest for nonproliferation and national security might arise from adaptation of the "conventional" physicist's bubble chamber. For example, reasonably high resolution imaging of fast neutrons should be possible by exploiting the ability to precisely locate the site of neutron interactions

within a bubble chamber. In addition, powerful active measurement systems could be constructed that exploit the temporal and spectral differences between interrogation and induced fission neutrons. Appendix A analyzes several possible approaches to this application. Consider, for example, the fact that a bubble chamber should be able to reject the AmLi neutrons used to induce fission in uranium with 99% efficiency while detecting induced-fission neutrons with roughly 50% efficiency. Further consider the fate of fast neutrons introduced into a room via a pulse neutron-generation system. Roughly 10 μs after the neutrons are introduced, they have either slowed to an energy below the detection threshold of the chamber or traveled so far away that return is highly unlikely. At this time, any fast neutrons recorded in the bubble chamber must inevitably have been the result of induced fission. Such an approach is expected to enable the searching of entire rooms for HEU.

A second but no less important application for BCs is as a low-cost medium for efficient neutron detection. Regardless of the working fluid, BCs do not require the use of expensive isotopes or electronics and thus may be low in cost. We believe that development of a dramatically lower cost neutron-detection technology (with comparable or improved efficiency and reliability) would function as an enabling technology for a variety of applications where a large number of neutron detectors is required. The efficiency of BCs is determined by the interaction probability of a neutron, the probability of bubble formation after an interaction, and the chamber's duty cycle. Since the first probability can be well over 50% for a several-inch-thick chamber, and the second probability is effectively 100%, the overall detection probability is determined primarily by the duty cycle. A research issue discussed in the next subsection involves determination of the conditions that will permit high-duty-cycle operation of BCs. Assuming that a duty cycle of 75% can be achieved, BCs may well exceed the efficiency of the conventional "moderate-and-capture" neutron detectors in use today.

4.3 Research Questions:

"Clean" Bubble Chambers: Research must be performed to determine the best method for developing a simple, low-cost bubble chamber with high efficiency. This chamber must use a working fluid that is acceptable from an environmental and safety point of view. Water has not been ruled out for this application, although heating to roughly 100°C would probably be required. Other relatively safe organic compounds should allow operation at temperatures closer to room temperature. A simple and effective method for pressure control is desired, preferably using piezoelectric crystals since they involve "non-mechanical" actuation. Finally, operating conditions that allow detection of neutrons within a "clean" chamber at a high duty cycle must be found. That is, the time during which the detector is active should equal or exceed the time during which it is recovering.

Bubble Chambers for Active Measurements: Research must be performed to optimize current BCs for use in active neutron measurements. This research would be designed to lead to a demonstration of the power of such an approach for applications like HEU search and HEU measurement. Appendix A discusses several approaches for the use of BCs for active measurements.

Boron Incorporation: Research is needed to determine the best way to introduce sufficient boron into droplet detectors similar to those commercially available today. There are several alternate approaches to achieving this. First, the Freon could be replaced with a boron-containing liquid that could be superheated at accessible temperatures and pressures. Second, a boron-containing material could be dissolved within the Freon. Both of these approaches assume that the boron can be largely kept out of the polymer matrix that holds the droplets. A third approach would entail adding boron to the polymer matrix while increasing the number density of droplets to the point that the nuclear-reaction products from within the matrix would be likely to strike at least one droplet. A high droplet density would require a high-droplet loading and/or the use of relatively small droplets. A final possible approach would be the

dispersal of a chemically inert, boron-containing powder such as boron carbide powder (B_4C) within the Freon droplets. However, preliminary tests of this approach indicate that the finely divided material forms nucleation centers. The introduction of lithium is a less attractive possibility since the products of the reaction ^6Li (n,t)^4He deposit less energy in the 100-nm distance of importance for bubble nucleation. Appendix B provides additional information about the issues involved in creating high-sensitivity droplet detectors.

Gradient Chamber: There is a type of bubble chamber that was explored around 1960 and subsequently rejected for use in particle-physics experiments. This detector has the advantages of relative simplicity and constant sensitivity in time. This approach uses a combination of gravity, temperature gradient, and concentration gradient to detect radiation interactions. Unlike conventional BCs, this detector requires no cyclic operation or pressure changes. The detector was rejected because of problems involved in achieving sensitivity to low linear-energy transfer (LET) radiation types such as muons, etc. The nuclear-recoil events resulting from neutron interaction should be far easier to detect and therefore may enable the construction of a viable gradient chamber.

Microchambers: We believe that very small BCs formed from glass tubes or capillaries should be explored as a novel approach to neutron detection. A large number of such microchambers operated together would have many of the advantages of BCs generally, but may not require rapid cyclic operation.

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Appendix A: Active Measurements

Appendix A: Active Measurements

This appendix analyzes three possible approaches to the use of bubble detectors for active measurement. The results described below are based upon Monte Carlo N-Particle Transport Code (MCNP) neutron-transport calculations and careful estimates of the efficiency with which energy-thresholded bubble chambers will record fast neutrons. The findings below establish our assertion that bubble detectors are an enabling technology that is ready for application for active measurements. The resulting measurement systems should lead to improved capability, reduced cost and complexity, and improved portability.

An active measurement involves looking for the neutrons resulting from induced fission. The most difficult part of any active measurement is finding a detection system for these induced-fission neutrons that does not respond to the "source" neutrons used for interrogation. Because of their ability to detect neutrons with effective time or energy thresholds, bubble detectors offer the ideal method for accomplishing active measurements. Furthermore, the cyclic operation of bubble chambers (see Section 3.0) is well suited for the use of pulsed neutron sources.

A summary of the findings below is given in advance:

- **Entire-Room Search**: A modestly sized bubble chamber set to record only neutrons above 200 keV in energy between 1 and 30 ms after a series of fast-neutron pulses allows detection of 5 kg of enriched uranium within a typical room with a diameter of 8 m. Such a measurement would be complete in only a few seconds. Uranium becomes much easier to detect in smaller rooms.

- **Container Characterization**: The same bubble chamber with a threshold set to 2.5 MeV would allow measurement of 1 kg of enriched uranium from a distance of 50 cm when a d-d neutron generator is used for interrogation. Such a system would be relatively portable and inexpensive compared to the "shuffler" and differential-die-away technologies that require a moderating "cave" to be placed around the container being interrogated. The measurement could be completed in substantially less than 1 min.

A.1 Entire-Room Uranium Search

In this application, a pulsed source of fast neutrons is used to interrogate an entire room for the presence of uranium. The logic is simple: any fast neutrons present after moderation of the initial pulse must necessarily be induced-fission neutrons. A detector used for this application must be able to

- reject the strong flux of moderated thermal neutrons with high effectiveness
- reject the strong initial pulse of fast neutrons with high effectiveness.

Moderating detectors have trouble in both of these areas, leading to the complex and expensive technologies such as the "shuffler" and the "differential-die-away" detector. Bubble detectors, in contrast, are completely insensitive to neutrons below a chosen energy threshold and to neutrons that reach the detector before a reduction of pressure creates a superheated condition.

MCNP calculations were run for the case of an 8-m-radius room containing 50 kg of uranium in the shape of a disk with 1-cm thickness. The walls of the room were assumed to consist of 2.5 cm of water. While water is clearly not a typical construction material, the moderation provided by this assumed wall is expected to be typical of construction materials such as wood, concrete, etc. The induced

fission signal will scale approximately linearly with the amount of uranium present since the uranium configuration is not close to criticality. Although the signal will depend upon the uranium geometry, the dependence should not be strong.

Figures A.1 and A.2 show the flux profile both with and without the uranium disk present for five energy bins. The selected bins are thermal (0 to 1 eV), low epithermal (1 eV to 1 keV), high epithermal (1 keV to 100 keV), lower-energy fast (100 keV to 1.0 MeV), and higher-energy fast (above 1.0 MeV). The curves show the number of neutrons striking the outer room wall normalized to the number of source neutrons in the initial pulse.

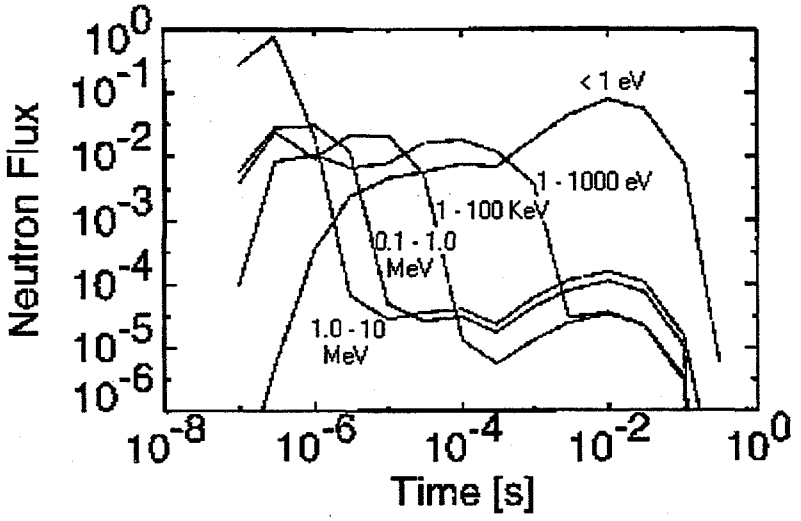

Figure A.1 Normalized Neutron Flux at the Room Wall as a Function of Time with a 50-kg Disk of Uranium Present. The five curves are labeled for different energy bins.

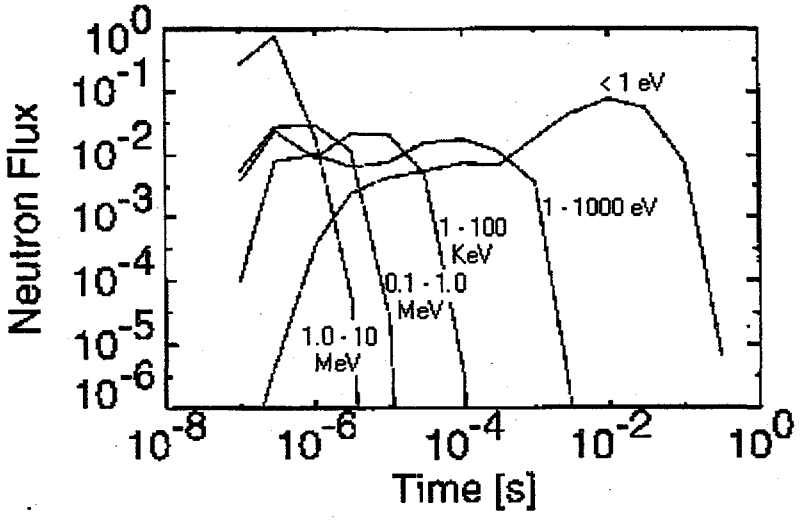

Figure A.2 Normalized Neutron Flux at the Room Wall as a Function of Time Without a 50-kg Disk of Uranium Present. The five curves are labeled for different energy bins.

Figures A.1 and A.2 clearly illustrate that only with uranium present are epithermal and fast neutrons present at late times. This is simply because the initial source neutrons *must* steadily moderate if

they are to remain in the room. Note that without uranium, the epithermal and fast neutron fluxes decrease effectively to zero at times between 10 μs and 3 ms after the original pulse. Epithermal neutrons persist longer in the room without uranium because of the longer time required for moderation. Further note that the thermal neutron flux depends little upon whether uranium is present. Neutrons derived from the original interrogation pulse dominate the thermal neutron flux at all times. These results are easy to understand in terms of the speed of a neutron. For example, a 1.0-MeV neutron travels at roughly 1.4 cm/ns. In 10 μs, therefore, a 1-MeV neutron has time to travel 140 m, or roughly 14 times across the diameter of this spherical room. Clearly, such a neutron must have collided numerous times with the materials in the wall to have remained in the room.

When uranium is present, neutrons of all energies are present until as late as 1 full second after the source pulse. This is the basis upon which a bubble chamber can infer the presence of uranium within the room. A bubble chamber can easily be set to record neutrons above a specific energy threshold. Upon consideration of Figures A.1 and A.2, a threshold of roughly 100 keV would seem to be a useful choice. In actuality, it is difficult to set an energy threshold below 200 keV. More than 50% of the induced fission flux above 100 keV occurs between 1 and 10 ms after the source pulse. A bubble chamber set to record fast neutrons between 1 and 10 ms after a source pulse is sensitive only to induced fission neutrons and should record virtually no neutrons from the interrogation source. The detectability of 5 kg of uranium can be calculated as follows. The normalized flux between 1 and 30 ms from 50 kg of uranium was 7×10^{-4}. Thus the flux from 5 kg would have been roughly 7×10^{-5}. Since these neutrons can strike anywhere on the outer wall, the flux per unit area becomes 3.5×10^{-11} cm^{-2} per source neutron. Assuming a 30% efficient detector with an area of 1000 cm^2, the number of detected neutrons becomes roughly 10^{-8} per source neutron. Assuming that 100 pulses with 10^9 neutrons each are used, the total number of detected neutrons becomes roughly 1000. This neutron emission level is easily achieved with a d-t generator and is close to becoming possible with commercial d-d generators. The number of background counts recorded during the same time period would have been roughly 13 neutrons at Hanford. Somewhat higher or lower backgrounds exist at other locations. We conclude, therefore, that 5 kg of uranium can be detected within an 8-m-diameter room is possible in only a few seconds under the assumptions outlined here.

Container Characterization

The uranium within a container could be characterized using a bubble chamber and a d-d neutron generator source. Unlike "shuffler" detectors and differential-die-away detectors, no moderator would be required. Fast-neutron–induced fission takes place within ^{235}U with a cross section of roughly 2 barns per atom at the 2.45 MeV energy of d-d neutrons. Suppose that a d-d generator capable of producing 100 pulses of 10^8 neutrons each is placed within 50 cm of the center of a container containing uranium. Again suppose that a detector with an area of 1000 cm^2 is available. For this application, an energy threshold of 2.5 MeV will ensure that interrogation neutrons are not detected. The expected efficiency with which fission neutrons are detected by such a detector is calculated to be 3.1%. Note that this is a spectrally weighted efficiency. MCNP calculations indicate that the spectrally weighted efficiency with which induced fission neutrons are created is roughly 2.8×10^{-6}. The overall efficiency with which neutrons are recorded normalized to the rate at which interrogation neutrons are emitted becomes 8.5×10^{-8}. If we assume that a single kg of uranium is present within the container, then an average of 85 neutrons would be recorded from 100 pulses of 10^7 neutrons each. The neutron background during this time should be very, very low since the time window during which neutrons are recorded is only 0.1 ms per pulse. We conclude, therefore, that detection of 1 kg of uranium within a container should be possible in only 10 to 20 seconds under the assumptions outlined here.

Appendix B: High-Sensitivity Droplet Detectors

Appendix B: High-Sensitivity Droplet Detectors

As discussed in the main text, the addition of a material such as ^{6}Li or ^{10}B to current droplet detectors should lead to a tremendous increase in sensitivity. A scaled-up version of such a detector would offer efficiency competitive with current large-area neutron detectors, but at substantially reduced cost. This appendix details some of the options and issues involved in the development of such detectors.

The physical requirement placed upon the developer is that the reaction products from the ^{6}Li and ^{10}B neutron capture reactions must deposit energy within the superheated droplets themselves. Materials containing the isotopes such as ^{6}Li and ^{10}B can be incorporated:

- into the plastic matrix
- into the droplet material
- into the droplet/matrix interface.

The neutron-sensitive isotope can be put into these

- by chemical incorporation
- by dissolving into the medium
- by physical mixing
- by incorporation into a surfactant.

One commercial vendor[3] has incorporated ^{6}Li into the plastic matrix and improved the thermal-neutron flux efficiency compared to the fast-neutron flux efficiency by a factor of greater than 20. In doing so, the thermal-neutron efficiency was increased by several orders of magnitude over that of conventional droplet detectors.

Despite this encouraging result, we believe that far more sensitivity enhancement is possible for two reasons. First, the commercial vendor used ^{6}Li instead of ^{10}B. The boron reaction has both a higher cross section and more highly charged products. The higher cross-section alone should increase thermal neutron sensitivity by a factor of roughly 5, whereas the boron reaction products will more easily lead to bubble nucleation within droplets. Further, the vendor placed the neutron capture agent, ^{6}Li, within the polymer matrix rather than at or within the droplets themselves. Clearly, many of the neutron-capture reactions will not deposit energy within the droplets, leading to lower efficiency.

Most organic compounds containing boron are toxic, flammable, or explosive or are extremely sensitive to water (or all three). These are not the ingredients for a low-cost neutron detector. One boron-containing compound of potential interest is carborane ($H_2C_2B_{10}H_{10}$), which is stable, insoluble in and unractive with water, and slightly soluable in nonpolar solvents. Two other attributes make it yet more suitable: first, it contains 10 boron atoms per molecule and second, there are two accessible hydrogen atoms that may be substituted to improve its solubility in the droplet material. Carborane is an article-of-commerce costing \$30/g to \$40/g for natural boron. Based on \$3.50/g for 95% enriched ^{10}B, the cost of enriched-boron carborane should be about \$55/g to \$60/g. It is considered to be toxic and potentially flammable.

We performed some simple experiments to determine if boron could be introduced into the matrix via suspension. Boron nitride powder and a boron-coated aluminum chip were introduced into commercial droplet detectors. The samples were exposed to thermal neutron fluxes and compared to

[3] Bubble Technology Incorporated

blanks and controls in which carbon powder was used in lieu of the boron nitride. The experiments revealed that the use of powder suspensions is a flawed approach because the particles provide surfaces on which heterogeneous nucleation of bubbles can occur. The boron-coated surface showed no evidence of significantly increased bubble formation. An improved option, fabrication of chemically modified droplet detectors, is beyond the scope of this exploratory work.

Table B.1 describes the relative merits of the possible options for incorporating ^{6}Li or ^{10}B into the droplet detector. The plastic matrix is typically a gel consisting of a polar material; the droplet material is typically a nonpolar Freon- or propane-like material. Note that the polymer matrix is hydrophilic while the droplets are hydrophobic. This allows for the possibility of adding thermal-neutron-sensitive matter into the detector with significant partitioning between the droplet and detector material.

Table B.1 Comparison of the Options for Incorporating ^{6}Li or ^{10}B into Droplet Detectors

	Into Plastic Matrix	**Into Droplet Material**	**Into Interface**
Chemical Incorporation	Lithium hydroxide may form gel suitable for matrix.	Nonpolar boron compounds may be grafted onto Freons or propane-like materials. Boron organics tend to be costly. Lithium is difficult to incorporate into nonpolar organics.	Lithum may be incorporated into soaps that will go to the droplet surface. These may lower the surface tension and make the detector less stable. Incorporated into the surface, either alpha or triton is likely to pass through droplet.
Solution	Lithium hydroxide can readily be added to the matrix, as it is hydrophilic. The oxide form of boron is slightly acidic and tends to react with basic gels.	Nonpolar boron compounds are soluable in nonpolar Freons and propane-like materials. Boron organics tend to be costly. Lithium is difficult to incorporate into nonpolar organics.	N/A
Physical Mixing	Mixing powdered form into matrix will provide a surface for heterogeneous nucleation.	Mixing powdered form into droplet will tend to cause heterogeneous nucleation.	N/A

It is possible to estimate the potential marginal cost and value of adding ^{10}B-containing materials to the bubbles in a superheated droplet detector. The assumptions for this analysis are

- the detector is 100 cm long by 25 cm wide by 0.5 cm thick
- the droplet loading is 10% (volume)
- the average bubble diameter is 25 μm
- the droplet material is Freon with a molecular weight of ca. 121
- the molar solubility of carborane is 1%
- the boron enrichment is 95%.

Based on these assumptions, the boron in the detector would be nearly 15% efficient in attenuating thermal neutrons. The cost of adding enriched boron to the detector would be ca. $150. Several such layers, used in conjunction with appropriate amounts of moderating material, would make possible an inexpensive, high-flux-efficiency neutron detector.

In reality, this efficiency is optimistic. Not all boron neutron-capture reactions will be detected; some recoils will be into the gel or otherwise ineffective. Also, the gel, which is typically an acrylamide polymer, will have some small solubility for the boron compound (as well as for the droplet material); as a result, some parasitic boron neutron-capture reactions will occur in the gel. In addition, the hydrogen in the gel will capture some neutrons. Nevertheless, the addition of ^{10}B to the droplet has the potential for significant improvement in performance at a reasonable cost.

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