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FACTORS AFFECTING THE RESPONSE OF THE BUBBLE DETECTOR BD-100 AND A COMPARISON OF ITS RESPONSE TO CR-39*

N. E. IPE, D. D. BURICK

*Stanford Linear Accelerator Center
Stanford University, Stanford, California 94305*

and

R. W. POLLOCK

Siemens Gammasonics, Des Plaines, IL 60018, U.S.A.

ABSTRACT

The BD-100 is a bubble detector available commercially from Chalk River Nuclear Laboratories, Canada for neutron dosimetry. According to the manufacturer, the BD-100 detects neutrons over an energy range of 100 keV to 14 MeV and the dose equivalent response is independent of energy. The sensitivity of the detector is dependent upon its temperature at the time of irradiation.

The sensitized detector self-nucleates upon sharp impact and when heated to temperatures of 48°C or greater. The BD-100 is insensitive to low energy gamma rays but responds to 6 MeV photons. The sensitivity (bubbles/μSv) of the BD-100 was found to be energy dependent when exposed to standard neutron sources with average energies ranging from 0.5 to 4.5 MeV. The bubbles formed upon irradiation continued to grow in size with time. The response of electrochemically etched CR-39 to the same neutron sources is also reported for comparison.

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INTRODUCTION

The BD-100 is a bubble detector available commercially from Chalk River Laboratory, Canada for neutron dosimetry. The detector consists of tiny superheated liquid droplets that are dispersed throughout a firm elastic polymer contained in a small sealed glass test tube.¹ A low boiling point liquid inside the detector tube exerts pressure on the polymer so as to keep the detector in a radiation insensitive state. The detector is sensitized by removing the plastic seal, unscrewing the cap and allowing the liquid to escape. Upon irradiation, neutrons interact with the detector medium, producing secondary charged particles. These charged particles interact with the superheated liquid droplets, causing them to vaporize, thus resulting in the formation of visible bubbles. The bubbles are trapped at the site of formation and the number of bubbles is a measure of the neutron dose-equivalent.

According to the manufacturer the BD-100 exhibits the following properties.²

- a) Detects neutrons over an energy range of 100 keV to 14 MeV
- b) Flat dose equivalent response with energy
- c) Detects thermal neutrons
- d) Insensitive to gamma radiation
- e) Isotropic response
- f) Lower detection limit $< 10 \mu\text{Sv}$

In addition the sensitivity of the detector increases as temperature of the detector increases.

In this paper the results of studies performed to evaluate some of the factors affecting the response of the BD-100 and some general observations regarding the

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detector are reported. The response of the detector to standard neutron sources with average energies ranging from 0.5 to 4.5 MeV is presented. Since CR-39 is also being used for fast neutron detection, its energy response under similar irradiation conditions is also presented for comparison.

PRINCIPLE OF OPERATION

Apfel's superheated drop detector^{3,4} and the BD-100 operate on the same principle, namely boiling in a superheated liquid can be initiated by the presence of secondary charged particles formed by radiation interactions with the detector. These charged particles deposit energy along their path, thus initiating the vapor bubble formation. The actual mechanism of vapor bubble nucleation is not fully understood but can be explained using Seltz's thermal spike model. According to this model, intense ionization and excitation along the path of the charged particle produces local heating, thus resulting in the formation of a minute vapor bubble. If a bubble reaches a size (defined by the critical radius) that makes it thermodynamically unstable, it will grow to a visible size by the evaporation of the superheated liquid until the whole droplet is consumed. The critical radius R_c is given by

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

where $\gamma(T)$ is the surface tension at temperature T and ΔP is the pressure difference between the vapor pressure of the superheated liquid droplet and the ambient pressure (i.e., pressure exerted on the droplet by the polymer). Thus ΔP is a measure of the degree of superheat. As ΔP increases R_c decreases and the energy or heat required for vaporization of the droplet is less. Thus the

more superheated a liquid is the less energy it needs to initiate bubble formation. The neutron energy threshold of detection depends upon the critical radius and the stopping power (i.e., the amount of energy deposited per unit path length) of the secondary charged particle. For a neutron of a given energy interacting with a given detector medium, the neutron energy threshold can be lowered by increasing ΔP .

EXPERIMENTAL METHODS

Three batches of detectors with sensitivities to PuBe neutrons of 0.198, 0.240 and 0.242 bubbles/ μ Sv at 20°C (within 10% as quoted by the manufacturer) were used in these studies. The bubble detectors were normally stored in the refrigerator and were taken out of the refrigerator about an hour before use. The detectors were sensitized as described earlier, and usually used within fifteen minutes after sensitization. According to Ing⁶ 100% of the sensitivity is obtained when the BD-100 is used within eight hours after sensitization. Immediately after irradiation electronic contact cleaner (containing freon) was introduced into the detector tube and the recapped detector was stored at room temperature. The detectors were usually counted about 15 hours or more after irradiation. This elapsed time period allowed the bubbles to grow to a reasonable size which facilitated the counting process described below. The magnified image of the bubble detector was projected on the T.V. screen with the aid of a video camera. A light source with variable intensity and orientation was used to obtain the best possible image on the screen. Each detector was rotated three times and the number of bubbles counted by scoring on a transparency sheet overlaying the T.V. screen. The highest reading for each detector was recorded.

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All neutron irradiations were performed outdoors in a low scatter surrounding with the detectors mounted on a water phantom. Corrections for neutron scattering and anisotropy of the neutron sources (except ^{252}Cf) were made in calculating the neutron fluences. The anisotropy correction for ^{252}Cf was not known. The fluences were converted to dose equivalents using the methods outlined in NCRP 70.⁶ The calculated values are reported. In general two to four detectors were used per irradiation condition. Variations as large as 35% were sometimes observed between detectors exposed to the same neutron dose equivalent. No differences in sensitivities were observed between the three batches of detectors.

The photon exposures were made with a Clinac 1800 medical accelerator. Three detectors mounted on a styrofoam stand were placed on the treatment couch such that the detectors were inside the primary beam in the patient plane. A field size of 35 cm $\times$ 35 cm was used. Two detectors similarly mounted were placed outside the beam at a distance of 50 cm from the isocenter in the patient plane.

The CR-39 used in this study was obtained from American Acrylics and Plastics, Inc., U.S.A. The detectors 1.587 cm $\times$ 2.85 cm $\times$ 0.063 cm in dimensions were covered with a polyethylene layer of thickness 0.01 cm. All irradiations were performed with the polyethylene intact. Two etching procedures were used. Procedure A consisted of an electrochemical etch in 6.5N KOH at 1500 volts, 60 Hz and 60°C for five hours, followed by a high frequency etch at 2 kHz for 23 minutes (blow-up step) and a 15 minute post etch in the oven to smooth the tracks for counting. Procedure B consisted of a similar etch but at a higher voltage of 3000 volts with time reduced to three hours.

RESULTS AND DISCUSSION

More detailed descriptions of factors that affect the response of the BD 100 are reported elsewhere.⁷

BACKGROUND

The average background on a sensitized unirradiated detector varied generally from about 0 to 3 bubbles. Hence no background subtraction was made in the number of bubbles reported for irradiated detectors.

FORMATION AND GROWTH OF BUBBLES

Bubble size distribution and concentration were non uniform within the detector with the concentration at the top of the detector medium being much greater than elsewhere within the detector. The growth rate was greatest the first day but growth continued for weeks.

EFFECT OF STORAGE TIME

There was no significant difference between the number of bubbles obtained for detectors exposed to a given neutron dose equivalent when counted one day and five months after irradiation. This indicated that the detector did not undergo any significant fading during this time period.

The detector medium of a sensitized unirradiated detector stored at room temperature without the cap started deteriorating after two weeks. Adding freon and recapping the detector prevents deterioration during storage.

EFFECT OF IMPACT

Irradiated detectors that were accidentally dropped on the floor one day after irradiation produced clusters of tiny bubbles in addition to existing ones as can be seen from Figure 1. The larger bubbles are due to neutron irradiation.

EFFECT OF PHOTON IRRADIATION

A sensitized detector exposed to 15 Gy of ^{60}Co gamma rays produced no bubbles. X-ray imaging and energy dispersant X-ray analysis of a small portion of the detector medium indicated that it has some high atomic number constituents. This makes the detector inherently self shielding at low gamma energies. In addition at these low energies the gamma rays interact mainly with the electrons and are not very efficient at depositing their energy locally.⁸ Thus the BD-100 is insensitive to low energy gamma rays. At higher energies (≥ 6 MeV) where the photon energy is comparable to the binding energy per nucleon, the probability of photo-nuclear interactions increases.

In order to study the effect of higher energy photons the bubble detectors were exposed inside and outside the primary bremsstrahlung beam produced by 6 MeV electrons. The exposures were made so that the detectors inside the primary beam received a dose of 10 Gy. The average number of bubbles and the standard deviation for the three detectors was 96 ± 7 . These bubbles are probably due to the protons generated by the photo-disintegration of the deuterium in the hydrogen which is a constituent of the detector medium. This reaction has an energy threshold of 2.22 MeV. Of the two detectors placed outside the primary

beam one detector had five bubbles, the other had none (when the total dose inside the primary beam was 20 Gy).

EFFECT OF TEMPERATURE

The sensitivity of the detector depends on ΔP which in turn is determined by the vapor pressure of the liquid droplets. As the temperature of the detector increases the vapor pressure increases, thus increasing the sensitivity. Temperature corrections for the BD-100 can be made using the temperature dependence curve provided by the manufacturer. However caution must be exercised in applying any temperature correction when the detector is irradiated in an environment at a higher ambient temperature than 20°C. This is because the BD-100 has an inherent thermal lag, i.e., the detector medium inside the tube takes some time to reach the ambient temperature when moved from an area where the ambient temperature is different. The changing sensitivity with temperature must be taken into account especially for short irradiation times.

The effect of temperature on sensitized unirradiated detectors can be seen in Figure 2. The first two detectors were heated in a water bath for about 45 minutes at approximate temperatures of 40°C and 48°C respectively. The third detector was left on the dashboard inside a closed car for about 11 hours when the outside temperature was about 28°C. This detector completely self-nucleated. Under such circumstances the dashboard can reach temperatures as high as 70°C. No bubbles were observed above background for the detector heated at 40°C. However, as is evident from the photographs, the BD-100 is thermally unstable when heated to temperatures of 48°C or greater.

LINEARITY OF RESPONSE

The response of the BD-100 was found to be fairly linear when exposed to PuBe neutrons in the dose equivalent range of 40 to 170 μ Sv. The limitation on linearity is governed to a large extent by the maximum number of bubbles that can be counted efficiently. Counting a large number of bubbles becomes difficult because of overlapping and the fact that the geometry of the detector does not lend itself to good resolution of individual bubbles.

ENERGY RESPONSE

The bubble detectors mounted on a water phantom were exposed to neutrons from standard sources such as PuBe, PuB, ^{252}Cf , PuF and PuLi, with average energies of 4.5, 2.3, 2.15, 0.9 and 0.5 MeV respectively. Figure 3 shows the sensitivity of the BD-100 as a function of average neutron energy. The average values are plotted at each energy. The error bars represent the standard deviations of the detectors. The curve is drawn only to aid the eye. The solid dots are the results of irradiations performed at ambient temperatures higher than 20°C (i.e., 35–40°C) and the triangles are for irradiations performed at an ambient temperature of about 20°C. The open circles represent the values obtained when the higher ambient temperature results are corrected for 20°C. The error bars for the open circles include an uncertainty of about 20% for the temperature corrections. The application of a temperature correction is not straight forward due to the fact that a) the detector has a thermal lag which depends on the difference between its temperature and the ambient temperature and b) there are variations in the ambient temperature during the period of irradiation. Some preliminary experiments indicate that it takes at least ten

minutes for the detector to reach a controlled ambient temperature of 30°C from its temperature of 19°C. However further refined studies are necessary. A thermal lag of 15 minutes was assumed in making the temperature corrections. The only conclusion that can be drawn from these results is that the response of the BD-100 appears to be energy dependent, the sensitivity generally increasing with decreasing neutron energy. The sharp increase in response to ^{252}Cf neutrons is not fully understood.

The sensitivity of CR-39 (expressed as the net track density/ μSv) as a function of average neutron energy is shown in Figure 6. The solid dots represent the results using etching procedure A and the open circles represent the results using etching procedure B. There does not appear to be any difference between the two procedures except at 4.5 MeV. Clearly CR-39 has a more pronounced dose equivalent energy dependence than the BD-100.

CONCLUSIONS

The BD-100 has many properties such as its high sensitivity, low threshold of detection, and isotropic response which make it a desirable neutron detector. However there are some factors that influence its response, such as temperature and impact which limit its use in the practical aspects of neutron dosimetry. The restriction of "use within eight hours after sensitization" and the difficulty of counting a large number of bubbles are additional handicaps. The BD-100 is also sensitive to high energy photons. In addition the detector has a response that appears to be energy dependent. Electrochemically etched CR-39 has a more pronounced energy dependence.

If most of the above mentioned problems can be overcome, the BD-100 would be a welcome addition to the field of neutron dosimetry.

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FIGURE CAPTIONS

- Fig. 1.** Effect of impact.
- Fig. 2.** Effect of temperature on sensitized unirradiated detector.
- Fig. 3.** Sensitivity of BD-100 as a function of average neutron energy.
- Fig. 4.** Sensitivity of CR-39 as a function of average neutron energy.

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Fig. 1

(a) ~40°C
(b) ~48°C
(c) ~48°C
Inside Closed Car

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Fig. 2

Fig. 3

Fig. 4