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BD–100: THE CHALK RIVER NUCLEAR LABORATORIES' NEUTRON BUBBLE DETECTOR*

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ABSTRACT

The BD–100 is a neutron bubble detector which is available commercially from Chalk River Nuclear Laboratories, Canada. The detector consists of tiny superheated liquid droplets that are dispersed throughout a firm elastic polymer. The polymer is contained in a small capped and sealed glass (glass detector) or plastic (plastic detector) test tube. A low boiling point liquid floating above the polymer exerts adequate pressure on it so as to keep it in a radiation insensitive state. The detector is sensitized by removing the cap and discarding the liquid. Upon neutron irradiation visible bubbles are formed in the detector, the number of bubbles being a measure of the neutron dose equivalent. One hundred percent of the sensitivity is obtained when the detector is used within eight hours after sensitization. The sensitivity is dependent upon temperature at the time of irradiation.

Studies performed with the glass detectors indicate that bubble size distribution and concentration are nonuniform within the irradiated detector. The bubbles continue to grow in size with time. Plastic and glass detectors exposed on an ICRU phantom to PuBe neutrons had sensitivities within 16 percent of the claimed values. The response of the glass BD–100 was fairly linear between neutron dose equivalents of 40 and 170 μSv.

Studies on the effect of storage time prior to and after irradiation showed that the detector did not undergo any fading for a period of five months. The sensitized detector is sensitive to impact and thermally unstable at temperatures of 48° C or greater. The BD–100 is insensitive to ^{60}Co-gamma rays but responds to photons of energies 6 MeV or greater. Results of irradiation in electron beams of energies 9 to 20 MeV are also presented. Exposure of detectors mounted on a phantom to neutron sources of average energies ranging from 0.5 to 4.5 MeV indicates that the dose equivalent response of BD–100 exhibits some energy dependence.

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1. Introduction

The BD–100 is a bubble detector which is commercially available for neutron dosimetry. The detector consists of tiny superheated liquid droplets that are dispersed throughout a firm elastic polymer.[1] The polymer is contained in a capped glass test tube which is sealed with a protective plastic coating. The total detector medium occupies a volume of 4 cm^3. A low boiling point liquid floating above the polymer exerts the appropriate vapor pressure so as to keep the detector in a radiation insensitive state. The detector is sensitized by cutting the protective coating, unscrewing the cap, removing the coating, 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. The number of bubbles in the detector is a measure of the neutron dose equivalent.

Figure 1 shows a sealed detector (a), a sensitized detector (b) and an irradiated detector (c).

According to the manufacturer, the BD–100 exhibits the following properties:[2,3]

- Detects neutrons over an energy range of 100 Kev—14 MeV
- Has a dose equivalent response independent of energy
- Detects thermal neutrons ($\frac{\text{Thermal neutron response in bubbles}/\mu Sv}{\text{PuBe neutron response in bubbles}/\mu Sv} = 0.076$)
- Insensitive to gamma rays
- Inherently isotropic response
- High detection efficiency
- Lower limit of detection $< 10\ \mu Sv$

- Can be custom made for neutron spectrometry.

In addition the detector is sensitive to temperature, its sensitivity increasing as the temperature increases.

Since the BD–100 is a fairly new detector in the field of neutron dosimetry, studies were performed to evaluate some of the factors affecting its response, including its energy response. In addition to these results, some general observations regarding the detector will also be reported in this paper.

2. Principle of Operation

The BD–100 and Apfel's superheated drop detector[4,5] operate on a principle very similar to that of the conventional bubble detector, namely boiling in a superheated liquid can be initiated by the presence of charged particles formed by radiation interactions with the detector. (The normal boiling point of a liquid is the temperature at which its saturation vapor pressure is equal to the external pressure. An increase in the external pressure results in an elevation of the boiling temperature and vice versa. The saturation vapor pressure depends on the temperature of the liquid, increasing as the temperature increases. A liquid can be raised to a temperature above its boiling point without boiling actually occurring. This metastable state of the liquid is known as the superheated state. The superheated state is very short lived, a slight disturbance or introduction of any impurities will cause the liquid to boil.) In the sensitized state (*i.e.*, by removing the excess liquid floating over the polymer), the external pressure has been reduced and the droplets are in a superheated state. Neutron irradiation results in the formation of secondary charged particles which deposit energy along their path, thus initiating the vapor bubble formation. The actual mechanism of vapor bubble formation is not fully understood but can be explained using Seitz's thermal spike model.[6] 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.*, the pressure exerted on the droplet by the polymer.) Thus ΔP is a measure of the degree of superheat. The more superheated a liquid is (*i.e.*, as ΔP increases) R_c decreases and the energy or heat required for vaporization of the droplet is less. The neutron energy threshold of detection thus depends upon the critical radius in addition to 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. Threshold detectors can be made for neutron spectrometry by appropriate choice of detectors with different ΔP.

The detection sensitivity (in bubbles/μSv) can be controlled by altering the amount of detector liquid dispersed in the polymer. It is our understanding that the detector liquid that is used is a combination of freons. The thermal neutron sensitivity can be attributed to the (n,p) reaction on chlorine and nitrogen present in the detector medium.[1]

3. Experimental Methods

Three batches of neutron detectors with sensitivities within 10% to PuBe neutrons of 0.198, 0.240 and 0.242 bubbles/μSv (according to the manufacturer) were purchased from Chalk River Nuclear Laboratories. All the detectors were stored in the refrigerator prior to use. The detectors were taken out of the refrigerator an hour before use, and used within 15 minutes after sensitization. All irradiations were performed with the cap of the detector removed. According to Ing[3] 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. The counting equipment is shown in Figure 2. The magnified image of the bubble detector (a) was projected on the T.V. screen (b) with the aid of a video camera (c). A light source (d) 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.

All neutron irradiations were performed outdoors in a low scatter surrounding. The detectors were mounted on a water phantom. Corrections for anisotropy of the neutron sources (except ^{252}Cf) and scattering of the neutrons were made in calculating the neutron fluences. The anisotropy correction for ^{252}Cf was not known at the time of irradiation. The fluences were converted to dose equivalents using the method outlined in NCRP 79.[7] All reported values of the dose equivalent are calculated. Measurements made with moderated BF–3 detectors indicated that the measured fluence rates were in reasonable agreement with the calculated values, except for ^{252}Cf and PuLi. The BF_3 measurements were higher than the calculated values by 16 percent for ^{252}Cf and 33 percent for PuLi. The moderated BF_3 detectors were calibrated with PuBe neutrons. In general two to four detectors were used per irradiation condition. Variations as large as 35 percent (from the large values) were sometimes observed between two detectors irradiated with neutrons under identical conditions. No significant 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 electron exposures were also made in a similar manner with the Clinac 1800. A field size of 15 cm $\times$ 15 cm was used. Two detectors were placed outside the beam at a distance of 37.5 cm from the isocenter.

4. Response of BD–100

BACKGROUND

The average background on a sensitized unirradiated detector usually varied from about 0 to 3 bubbles per detector during the first eight hours after sensitization. Hence no background subtraction was made in the number of bubbles reported for irradiated detectors.

FORMATION AND GROWTH OF BUBBLES

Bubble formation in the irradiated detector was nonuniform with the concentration at the top of the detector usually being much greater than elsewhere in the detector. There was also a large distribution in the size of the bubbles. The bubbles that were formed immediately upon irradiation were very small but they continued to grow in size with time after irradiation, as shown in Figure 3. The bubble growth rate reached a maximum within a day but growth continued for weeks. Most of the detectors had been sprayed inside with electronic contact cleaner immediately after irradiation. The basis for this was the assumption that the introduction of a low boiling point liquid inside the detector tube would restrict the bubble growth because of the pressure that it would exert on the polymer. However, no visible difference was found in bubble growth between detectors that had contact cleaner added inside the tube after irradiation and those that did not have the contact cleaner. The bubbles in the detector with no contact cleaner grew much larger in size over a given time period when the bubble density was low, as compared to when it was high. In the latter case the bubbles tended to grow into each other and coalesce because of overcrowding.

Later discussions with Ing[3] revealed that the introduction of freon 114 in the detector would reduce bubble growth because the vapor pressure that it exerts is adequate to restrict the growth. Both the electronic contact cleaners that we used contained freons other than freon 114.

COMPARISON WITH THE PLASTIC BD–100 DETECTOR

The BD–100 detectors are now available in plastic tubes (instead of glass) but with higher sensitivites ranging from 1.2 to 1.8 bubbles/μSv. The plastic detectors have a larger detector volume than the glass detectors. In order to compare their responses, a group of plastic and glass detectors were exposed on an ICRU phantom to PuBe neutrons so that the plastic detectors received a dose equivalent of 42 μSv and the glass detectors received a dose equivalent of 260 μSv. The temperature at the time of irradiation was 21.5° C. The results are summarized in Table 1. The sensitivities in Column 3 were corrected for temperature.

The glass detectors were much easier to count than the plastic detectors because of the following reasons. The glass detectors provided greater optical clarity than the plastic detectors. The plastic detectors were much larger in diameter, thus making the resolution of individual bubbles difficult, especially in the case of overlapping bubbles.

LINEARITY OF RESPONSE

The linearity of the response of the BD–100 was examined by exposing detectors to neutrons from PuBe with dose equivalents ranging from about 40 to 170 μSv. The results are shown in Figure 4. Two detectors were used at each dose equivalent and the individual detector response is plotted at each dose equivalent. In one case, one out of the two detectors proved to be defective. The outdoor temperature during these irradiations was about 26° C; however, all irradiation time periods were rather short, the longest being 12 minutes. The response at 166 μSv will have the largest temperature correction while the response at 41 μSv will have the smallest temperature correction (see Effect of Temperature). The response is relatively linear within experimental limits in this dose-equivalent range. The limitation on linearity at higher dose-equivalents will be governed to a large extent by the maximum number of bubbles that can be counted efficiently. For a large number of bubbles in the detector, the counting becomes difficult as the bubbles tend to overlap and the geometry of the detector does not lend itself to good resolution of the individual bubbles. An optimum number of bubbles for our counting purposes would lie somewhere in the range of 60 to 75.

EFFECT OF STORAGE TIME

Detectors that were stored in the refrigerator showed no significant loss of sensitivity when exposed on an ICRU phantom after five months of storage to PuBe neutrons. The average sensitivity for two detectors was found to be within ten percent of the expected sensitivity.

There was also 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, respectively. This indicates that the detectors did not undergo any appreciable fading during this time period. Occasionally we found that the bubbles which were formed very close to the walls of the tube would creep up against the walls, flatten out and eventually disappear with time.

Figure 5 shows a sensitized unirradiated bubble detector that was stored at room temperature without the cap for about 24 days. The polymer in the detector deteriorated after two weeks. Adding freon and storing the detector with the cap screwed on prevented deterioration.

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 6. The larger bubbles are due to neutron irradiation.

EFFECT OF PHOTON IRRADIATION

The BD–100 was found to be insensitive to gamma rays from Co-60 for doses up to 15 Gy. X-ray imaging and energy dispersant X-ray analysis of a small portion of the detector indicated that the detector is made up of some high atomic number materials such as cesium, chlorine and aluminum. This makes the detector inherently self-shielding. Also at these low energies the gammas interact with the electrons and do not efficiently deposit their energy locally.[6] The probability of photonuclear interactions increases at higher energies ($\geq$ 6 MeV) where the photon energy is comparable to the binding energy per nucleon.

In order to study the effect of higher energy photons, the bubble detectors were exposed inside and outside the primary bremsstrahlung beam produced by both 6 MeV and 15 MeV electrons, respectively. In the former case, exposures were made so that the detectors inside the beam received a dose of 10 Gy. The detectors outside the beam were exposed for a longer period (*i.e.,* the total dose inside the beam was 20 Gy). In the case of the 15 MeV beam, exposures were made so that the detectors inside the beam received a dose of 0.1 Gy. The results of these exposures are shown in Table 2. For the 6 MeV exposure, the bubbles produced in the detectors inside the beam are probably due to the protons generated by the photodisintegration of the deuterium in the hydrogen in the detector medium. This reaction has an energy threshold of 2.22 MeV.

It is well known that electron accelerators operating above 10 MeV produce significant quantities of photoneutrons,[7] so some of the response at 15 MeV can be attributed to the photoneutrons. Neutron fluences were measured at 15 MeV with moderated gold foils. These moderators have a response nearly independent of energy in the range of .03–4 MeV. A comparison of the response (on a fluence basis) of the BD–100 to that of moderated gold foils is shown in Table 3. While most of response shown is due to neutrons photon-induced reactions in the BD–100 are also possible. The following reactions are probable:

$$^{12}C(\gamma,\alpha)^{8}Be \rightarrow 2\alpha \quad \text{(threshold energy = 7.4 MeV)}$$

$$^{2}D(\gamma,n)p \quad \text{(threshold energy = 2.22 MeV)}.$$

However, since the exact constituents of the detector medium are not fully known, all the possible reactions cannot be listed.

EFFECT OF ELECTRON IRRADIATION

All electron exposures were made with a Clinac 1800, which provided electron energies of 6, 9, 12, 16 and 20 MeV. No bubbles were observed above background for detectors exposed to 6 MeV electrons, but at higher energies bubbles were produced, as can be seen from the results in Table 4. The exposures were made at a dose rate of 4 Gy/min. These bubbles could either be due to some neutron contamination in the primary beam, a possibility that has not been entirely ruled out, or due to the scattering of nuclei in the detector medium by the relativistic electrons. Further studies are underway in order to investigate the true cause of the response of these detectors.

EFFECT OF TEMPERATURE

The sensitivity of the detector depends upon ΔP which in turn is determined by the vapor pressure of the liquid droplets. As the temperature increases, the vapor pressure increases, resulting in an increase in sensitivity. Temperature corrections for the BD–100 can be made using the temperature dependence curve provided by the manufacturer (Figure 7); 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. Some preliminary studies 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.

The effect of temperature on sensitized unirradiated detectors can be seen in Figure 8. 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. The dashboard material is such that under these 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 figure, the detector began to self-nucleate when heated to temperatures of 48° C or greater. Thus the detector can only be used in environments where the ambient temperature is less than 48° C.

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. Figures 9a and 9b show the sensitivity of the BD–100 in bubbles/μSv and bubbles/n-cm^{-2}, respectively, 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 (the outdoor temperature varied from 35 to 40° C during these irradiations). The triangles are the results of irradiations performed when the outdoor temperature was 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 percent for temperature corrections. The application of a temperature correction is not straightforward 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. 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 dose equivalent response of the BD–100 appears to be energy dependent, the sensitivity generally increasing with decreasing neutron energy. The increased response to ^{252}Cf is not fully understood. The anistropy correction was later determined to be about 11 percent. The response of the

BD–100 on a fluence basis appears to have less of an energy dependence.

5. Conclusions

The BD–100 has many properties, such as its high sensitivity, low threshold of detection and isotropic response, that make it a desirable neutron detector. However there are some factors that influence its response, such as impact and temperature which limit its use in the practical aspects of neutron dosimetry. An ideal dosimeter should be rugged and insensitive to variations in the environmental conditions. The restriction of "use within eight hours after sensitization" and the practical difficulty associated with counting a large number of bubbles by eye are additional handicaps. The BD–100 is also sensitive to high energy photons ($\geq$ 6 MeV) and this limits its use around medical accelerators. In addition the detector has a dose equivalent response that appears to be energy dependent.

It is our understanding that some of the problems associated with the BD–100 are in the process of being resolved. According to the manufacturer, a workable method of temperature compensation between 20 and 40° C has been proven in the lab at Chalk River. It is also likely that reusable detectors which have an extended lifetime of one month after sensitization (if used continuously) and for three months (if recycled daily) will be available in the near future. Reusable plastic threshold detectors are now available for neutron spectrometry with energy thresholds of 0.01, 0.1, 0.5, 1.5, 3 and 10 MeV. The response of these detectors are now being investigated at SLAC. In addition SLAC is also in the process of designing an automatic counter for the BD–100 which detects each individual "pop" as the bubbles form upon irradiation, and converts it to an electrical signal. The processed signals are then counted. Preliminary tests indicate that the counter works fairly well. More detailed tests are currently being conducted.

ACKNOWLEDGMENTS

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Table Captions

Table 1. Response of glass and plastic detectors to PuBe neutrons.

Table 2. Response of BD–100 to 6 and 15 MeV bremsstrahlung.

Table 3. Comparison of response of BD–100 and moderated gold foils to 15 MeV bremsstrahlung.

Table 4. Effect of electron irradiation.

Table 1

Detector	No. of Detectors	Sensitivity (Bubbles/μSv)	Dose Equivalent (μSv)	Average No. of Bubbles $\pm 1\sigma$	Expected No. of Bubbles
Glass	2	0.282	260	64 ± 4	73
Glass	3	0.310	260	89 ± 1	81
Plastic	2	1.818	42	88 ± 3	76

Table 2

Energy (MeV)	Dose Inside Primary Beam (Gy)	No. of Bubbles $\pm 1\sigma$ Inside Beam	Outside Beam	No. of Bubbles $\pm 1\sigma$ per Gy Inside Beam	Outside Beam
6	10	96 ± 7		9.6 ± 0.7	
	20		2.5 ± 3.5		0.12 ± 0.17
15	0.1	142 ± 14	36 ± 11	1420 ± 140	360 ± 110

Table 3

Detector	Neutron fluence per Gy ($n/cm^2 - Gy$) Inside Beam	Outside Beam
BD–100*	$(1.58 \pm 0.16) \times 10^7$	$(4.0 \pm 1.2) \times 10^6$
Moderated Gold Foils	1.13×10^7	3.86×10^6

* A conversion factor of 9×10^{-5} bubbles$/_{n-cm^{-2}}$ (taken from Figure 9b) was used.

Table 4

Energy (MeV)	Dose Inside Beam (Gy)	No. of Bubbles Inside Beam	No. of Bubbles Outside Beam	Av. No. of Bubbles $\pm 1\sigma$ Inside Beam	Av. No. of Bubbles $\pm 1\sigma$ per Gy Inside Beam
9	20	16	1	22 ± 5	1.10 ± 0.25
		23	0		
		26			
12	1	197		219 ± 21	219 ± 21
		239			
		222			
	6		4		
			6		
16	0.1	75	4	75 ± 10	750 ± 100
		65	3		
		86			
20	0.05	61	1	80 ± 21	1600 ± 420
		102	3		
		77			

Figure Captions

Figure 1. The BD–100 Bubble Detector.

Figure 2. Counting equipment.

Figure 3. Bubble growth with time after irradiation.

Figure 4. Number of bubbles as a function of neutron dose equivalent.

Figure 5. Effect of storage without cap.

Figure 6. Effect of impact.

Figure 7. The temperature dependence of BD–100 as provided by the manufacturer.

Figure 8. Effect of temperature on sensitized detector.

Figure 9. Sensitivity of BD–100 as a function of average neutron energy.

11-87 5853A1

Fig. 1

11-87

5853A2

Fig. 2

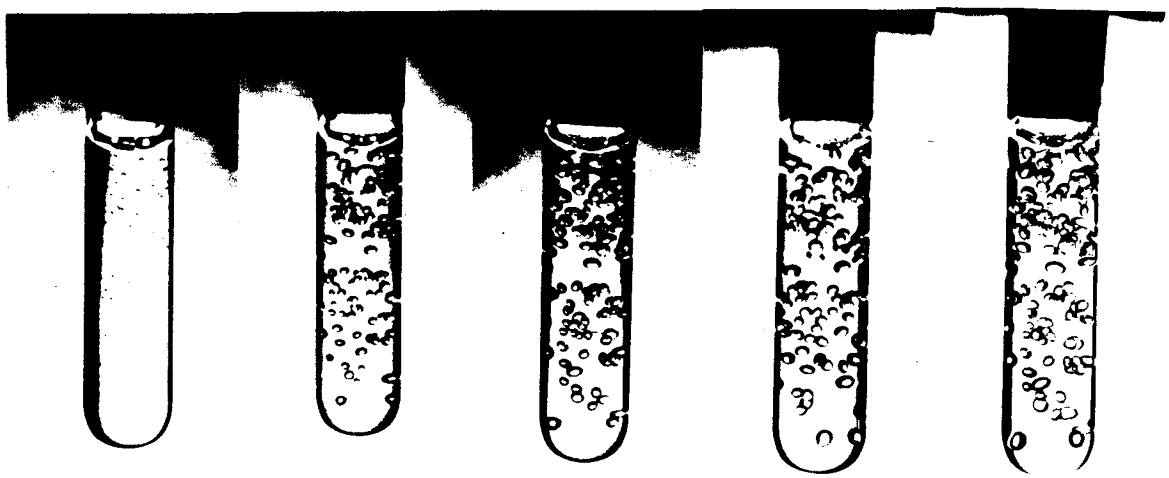


Fig. 3

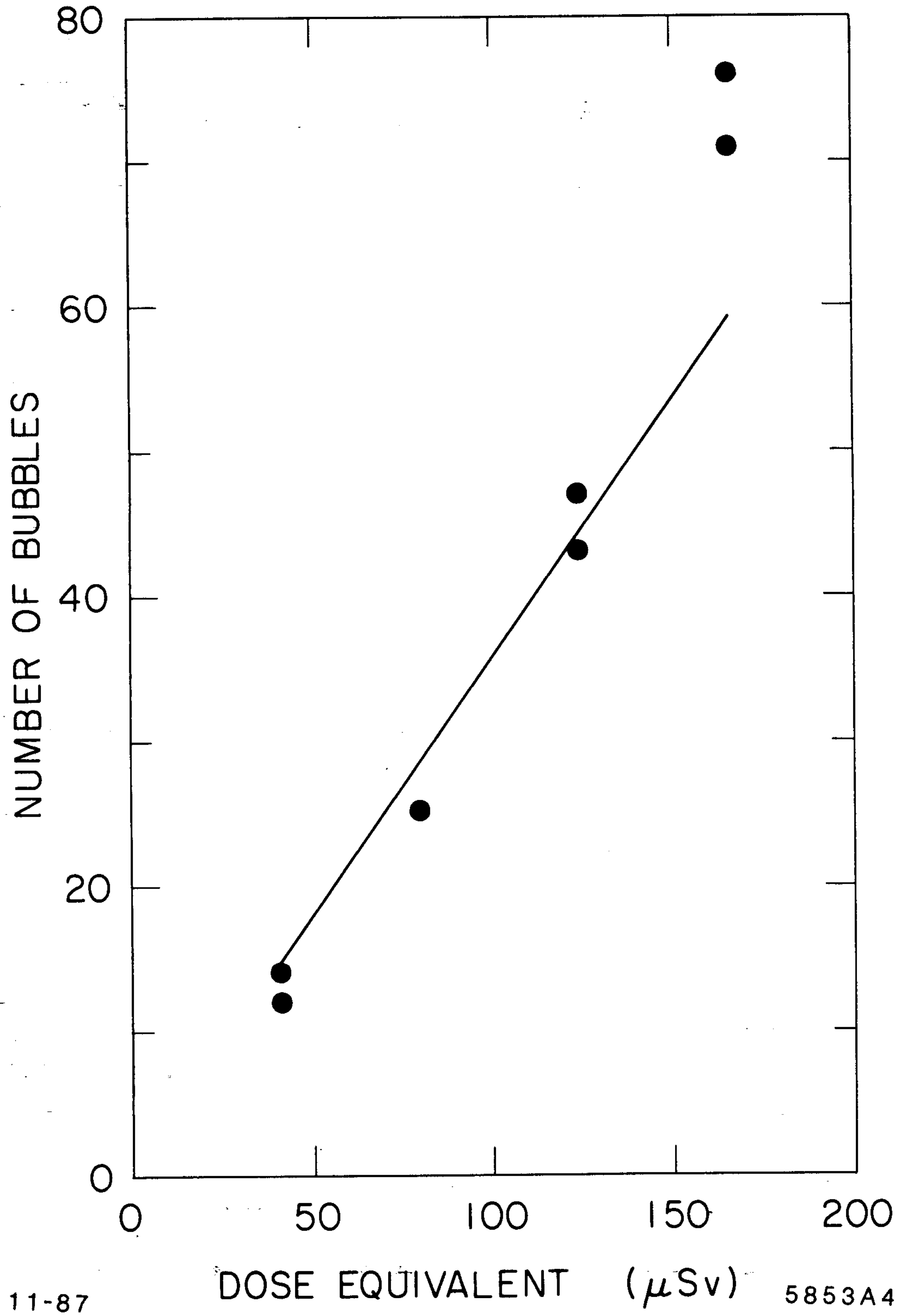

80
60
40
20
0
NUMBER OF BUBBLES
0
50
100
150
200
DOSE EQUIVALENT (μSv)
11-87
5853A4

Fig. 4

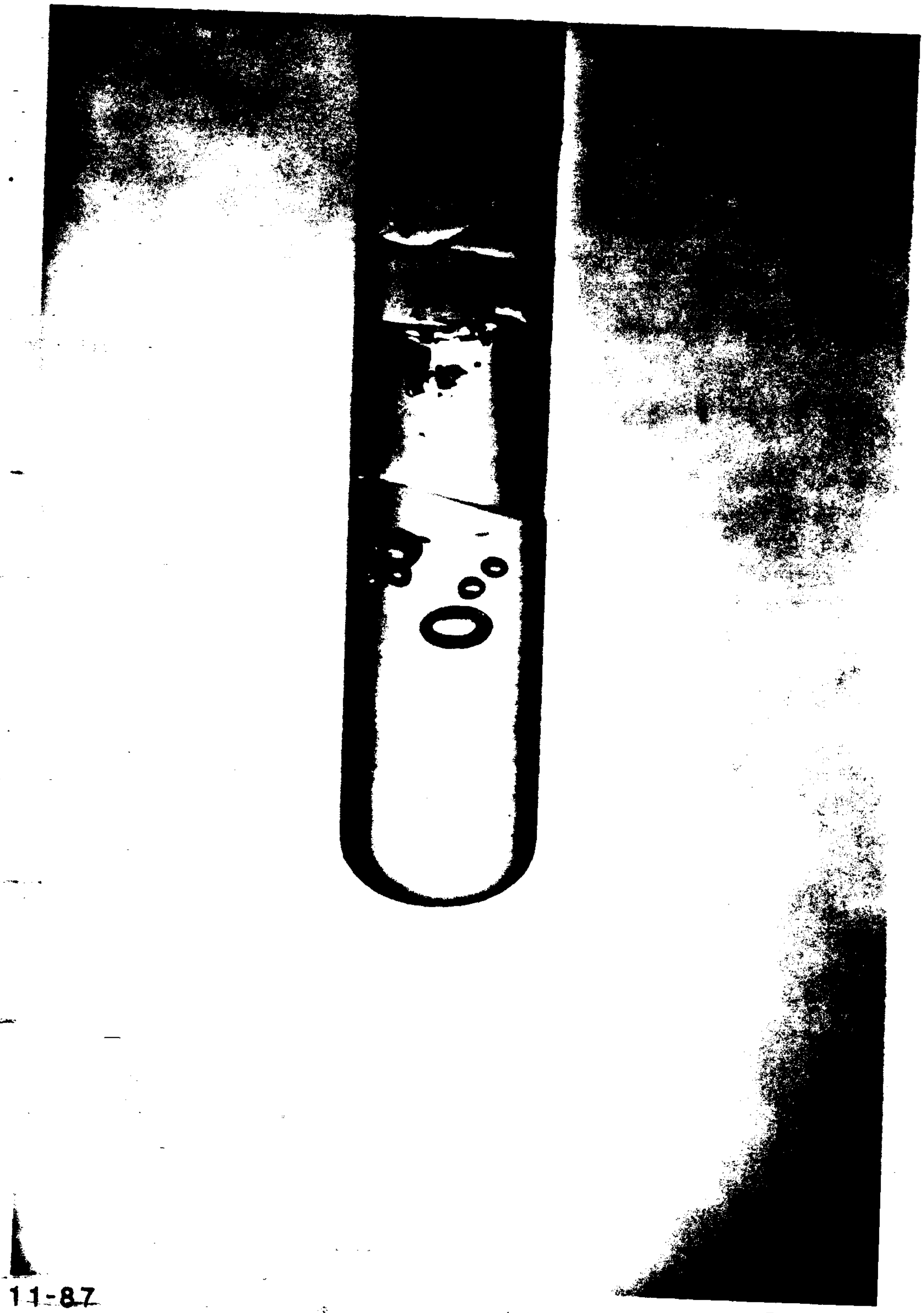
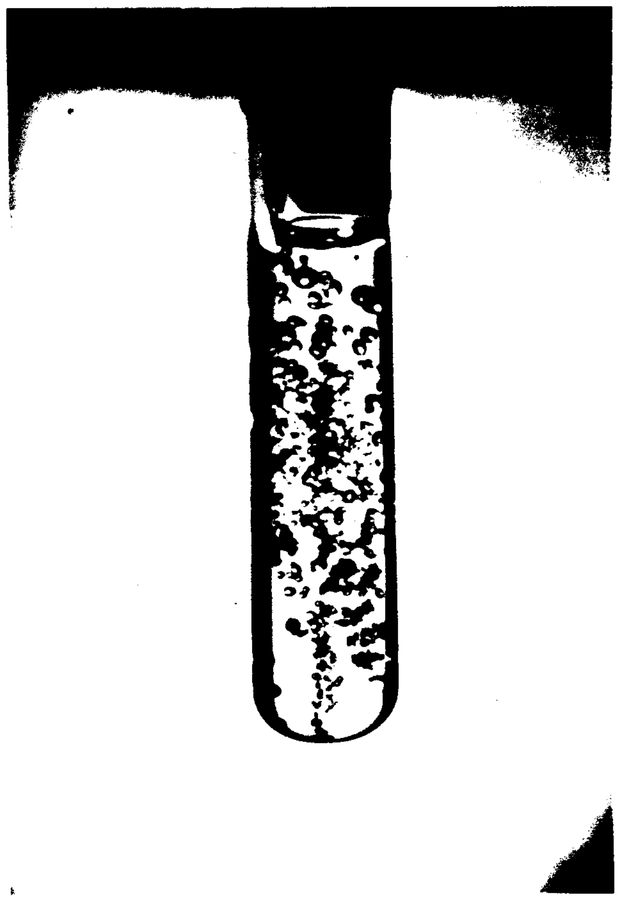
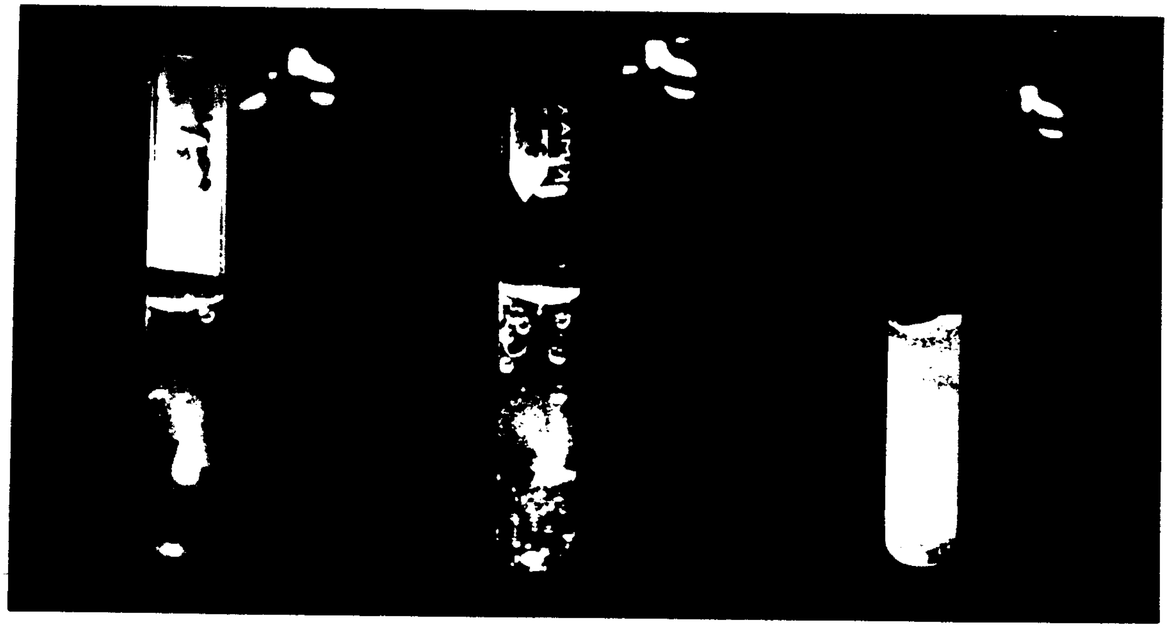


Fig. 5



9-87 5841A7

Fig. 6



(a) ~40°C

(b) ~48°C

(c) Inside Closed Car

9-87

5841A6

Fig. 7

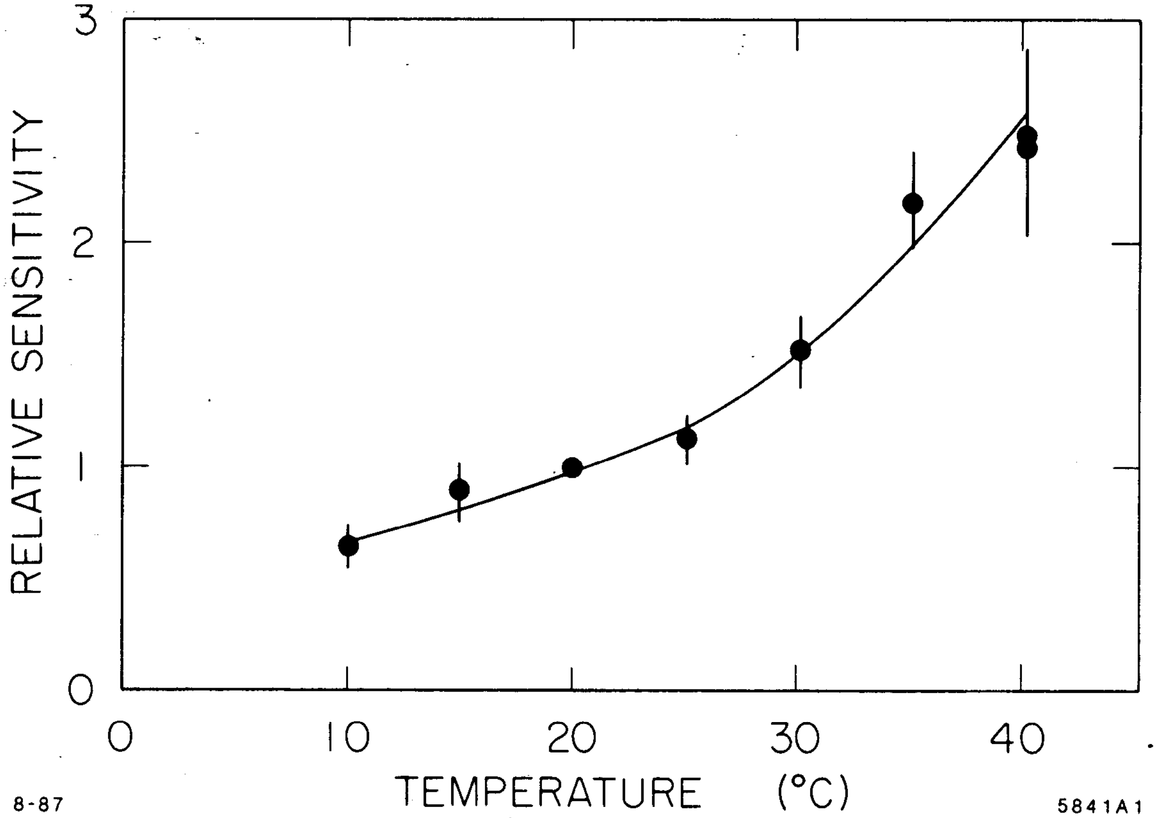
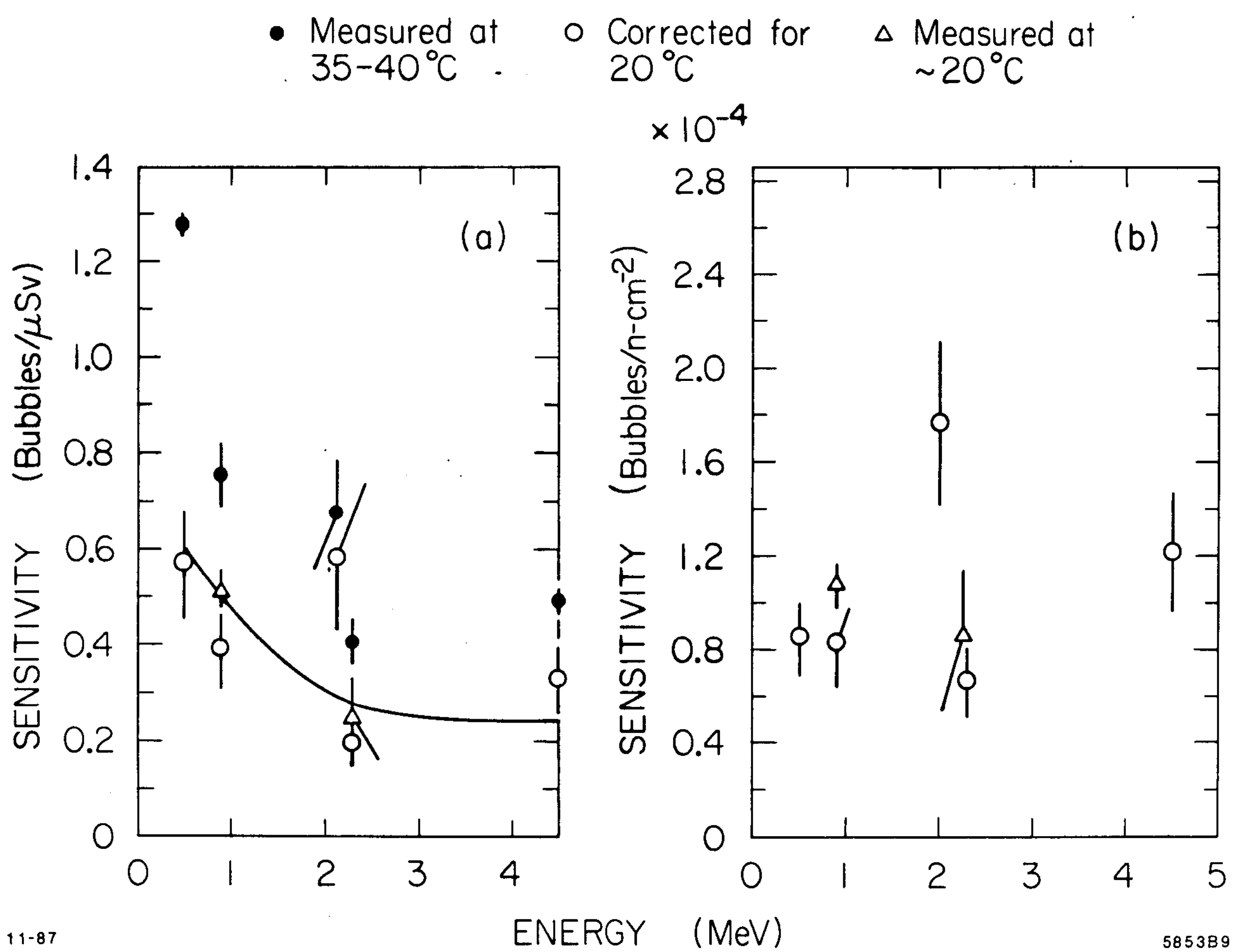

RELATIVE SENSITIVITY
3
2
1
0
0
10
20
30
40
TEMPERATURE (°C)
8-87
5841A1

Fig. 8



● Measured at 35-40°C
○ Corrected for 20°C
△ Measured at ~20°C
(a)
(b)
SENSITIVITY (Bubbles/μSv)
SENSITIVITY (Bubbles/n-cm^{-2})
× 10^{-4}
ENERGY (MeV)
11-87
5853B9

Fig. 9