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CHARACTERISTICS OF A RADON DIFFUSION CHAMBER WITH ELECTRICAL COLLECTION USING PLASTIC NUCLEAR TRACK DETECTORS

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BUREAU OF MINES
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A portable radon diffusion chamber has been tested with electrical collection of the radon daughter nuclei for enhancement of sensitivity. The detector size was small enough to be worn suspended from the belt. With a battery-supplied high voltage, the sensitivity was found to be 1.43 tracks/cm^2 per pCi-hr/ℓ of ambient radon at 50% relative humidity. The track densities on the surfaces of the CR-39 plastic nuclear track detectors were found to be non-uniform, making difficult the determination of average densities with high accuracy. The fabrication of electrets for use in the detector was investigated in accordance with published techniques with negative results.

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FOREWORD

This report was prepared by the University of San Francisco, San Francisco, California, 94117, under USBM Contract No. J0123037. The contract was initiated under the Metal and Non-Metal Health and Safety Research Program. It was administered under the technical direction of the Denver Mining Research Center with Mr. Robert Droullard acting as the Technical Project Officer. Mr. Doyne W. Teets was the contract administrator for the Bureau of Mines.

This report is a summary of the work recently completed as part of this contract during the period March, 1982, to November, 1983. This report was submitted by the authors in September, 1983.

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CHARACTERISTICS OF A RADON DIFFUSION CHAMBER WITH ELECTRICAL COLLECTION USING PLASTIC NUCLEAR TRACK DETECTORS

by

Allen L. Frank and Eugene V. Benton

ABSTRACT

A portable radon diffusion chamber has been tested with electrical collection of the radon daughter nuclei for enhancement of sensitivity. The detector size was small enough to be worn suspended from the belt. With a battery-supplied high voltage the sensitivity was found to be 1.43 tracks/cm^2 per pCi-hr/ℓ of ambient radon at 50% relative humidity. The track densities on the surfaces of the CR-39 plastic nuclear track detectors were found to be non-uniform, making difficult the determination of average densities with high accuracy. The fabrication of electrets for use in the detector was investigated in accordance with published techniques with negative results.

INTRODUCTION

In past studies, the University of San Francisco has investigated the use of plastic nuclear track detectors (PNTDs) in detecting radon-222 and its airborne daughter products (Frank and Benton, 1977; Frank et al., 1978; Frank and Benton, 1982). The major purpose of these studies was the development of a Working Level dosimeter for uranium miners. The approach to dosimetry which proved to be most practical involved simultaneous measurements with two PNTDs. One was exposed to the ambient air in order to measure the total airborne ^{4}He-particle activity. The other was enclosed in a diffusion chamber and measured only the radon activity. Together they gave total radon and daughter alpha activities plus a partial measure of the equilibrium ratio between radon and daughters. The problems encountered with this method of dosimetry were an expansion of the statistical and experimental errors when Working Level was calculated from the two measured alpha particle track densities, due to uncertainties in the individual daughter equilibrium values, and a susceptibility to contamination errors in an operational uranium mine.

The radon diffusion chambers (RDCs) used with the dosimeters were cylindrical, with cavities of 1.9 cm in diameter by 2.54 cm in length, and rectangular, with cavities 2.54 cm by 2.30 cm by 1.27 cm. The sensitivities of these detectors is 0.075 and 0.085 tracks/cm^2-hr per pCi/ℓ of ambient radon, respectively. The development of an RDC with enhanced sensitivity was undertaken to extend the useful exposure range from $\sim 10^4$ pCi-hr/ℓ down to $\sim$ 500 pCi-hr/ℓ. A detector of this sensitivity, and small enough to be carried routinely, could be used in a variety of circumstances to provide accumulated radon exposures to persons or in certain areas. An upper limit to Working

Level exposures could be determined or, if the Working Level ratios were known or estimated, then Working Level exposures would be determined by the radon exposures.

SENSITIVITY ENHANCEMENT

The sensitivity of the RDCs can be increased by enlarging the chamber size and by electrical collection of the charged daughter nuclei. A size change alone is not satisfactory, since the increase in sensitivity is sub-linear to increase in size, and response fluctuations with air density changes become a problem. A combination of increased chamber size with electrical collection of the daughters can greatly enhance the response.

Electrical collection has been tested in other studies. Of these, George (1977) used a chamber of 1.5ℓ volume with a 900 volt battery and a TLD detector; Kotrappa et al. (1981a, 1981b) used a 10ℓ chamber with an electret of approximately 3000 volts equivalent and an alpha scintillation system for counting the collected daughter nuclei. They also used both TLDs and PNTDs. Frank and Benton (1982) tested a chamber of 7.4 cm^3 volume with collection potentials up to 50 volts and with PNTDs to detect the alpha particles. Of the detectors tested, TLDs can accumulate a radiation dose over long exposure periods and are convenient to read out but are sensitive to gamma- or X-rays present. The PNTDs can accumulate particle tracks over long exposure periods and are insensitive to high energy photons, although the readout is more time-consuming. The scintillation detection method requires that a thin plastic film onto which the daughter nuclei have collected be removed from the chamber and placed in the scintillation instrument. Exposure times are limited to three to four hours and nuclei decay must be corrected for. Minimum detectable limits reported were 0.03 pCi/ℓ of radon for a one-week exposure (George, 1977), 0.03 pCi/ℓ for a three-hour exposure and scintillation counting (Kotrappa et al., 1981a and 1981b) and about 5 pCi/ℓ for a one-week exposure (Frank and Benton, 1982). For a one-week exposure and using TLDs and PNTDs, Kotrappa et al. (1981a) would have minimum detectable limits of about 0.01 pCi/ℓ of radon, as calculated from their reported sensitivities. The minimum detectable limit for a one-week exposure on the projected USF detector is about 0.3 pCi/ℓ of radon, where the size is small enough for routine carrying.

ELECTRICAL COLLECTION VOLTAGE

For a portable RDC the electrical collection voltage can be supplied only by battery or an equivalent, such as the electret. The voltage required is hundreds of volts (for chamber volumes of a large fraction of a liter) but the power requirements are minimal since the collection of the charged daughter nuclei represents an unmeasurably small current. Electrets appear to be the most satisfactory method of applying the voltage. Since they are only thin sheets of Teflon plastic, they are both small and light in weight. Batteries are also sufficient but are heavier and larger.

ELECTRET FABRICATION TESTING

Two of the methods were investigated. These were the liquid contact method and the heated-sample method. In the liquid contact method, a charging liquid is used between the electret material and one of the two electrodes which establish a charging voltage across the electret. The charging apparatus is described in Figure 1. In the condition shown, the upper electrode, which has the absorbent pad attached to it, is the positive electrode. It may be either positive or negative in electret production. The charge separation within the charging liquid is indicated, showing that positive charges are collected onto the top surface of the Teflon. The metallized bottom surface of the Teflon becomes negative. This is basically the apparatus described by Chudleigh (1976) and by Kotrappa et al.(1982). At the end of the charging period, Chudleigh slid the upper electrode and pad away from the charging area horizontally while Kotrappa et al. raised them vertically off the Teflon. They reported that stable charges were deposited onto their Teflon sheets which yielded voltages near the charging voltages used. Equal and opposite charges were retained on the two Teflon surfaces.

The heated-sample method of electret production is described by Kotrappa et al. (1980, 1981a). The metallized sheet of Teflon is clamped between two electrodes and placed in an oven at elevated temperature under a charging voltage. After the charging period, the Teflon is allowed to cool with the charging voltage still applied. This method was reported to yield electrets with voltages of 3 kV for charging voltages of 4.5 kV.

The liquid contact method of production was tried first since it appeared quite simple in scope. Kotrappa et al. (1980) reported that they used polytetraflouroethylene (Teflon TFE) while Chudleigh (1976) used Teflon FEP. We tested both materials. The Teflon TFE used was 0.08 cm thick, just as that used by Kotrappa et al. The Teflon FEP used was 250 μm while that used by Chudleigh was 25 μm. All of these material thicknesses should be applicable for electret production.

The metallized layer was applied to the Teflon by painting a lightly buffed area with colloidal silver. Areas of 1 inch in diameter were metallized since this matched the size of the upper electrode and was the electret size desired. The method of metallization does not appear to be important since vacuum evaporated aluminum (Chudleigh, 1976) and painted-on colloidal graphite (Kotrappa et al., 1982) have been used. The important consideration is that the lower surface of the potential electret be in intimate contact with the bottom electrode. Otherwise the charge distribution over the electret is spotty and the total charge is reduced.

A variety of charging voltages and charging times were tested, from 1000 to 6000 volts and for times of 5 to 30 minutes. Chudleigh (1976) studied these variables extensively, up to 2000 volts, for his 25 μm Teflon. Kotrappa et al. (1982) reported 2000 and 5000 volts used with their 0.08 and

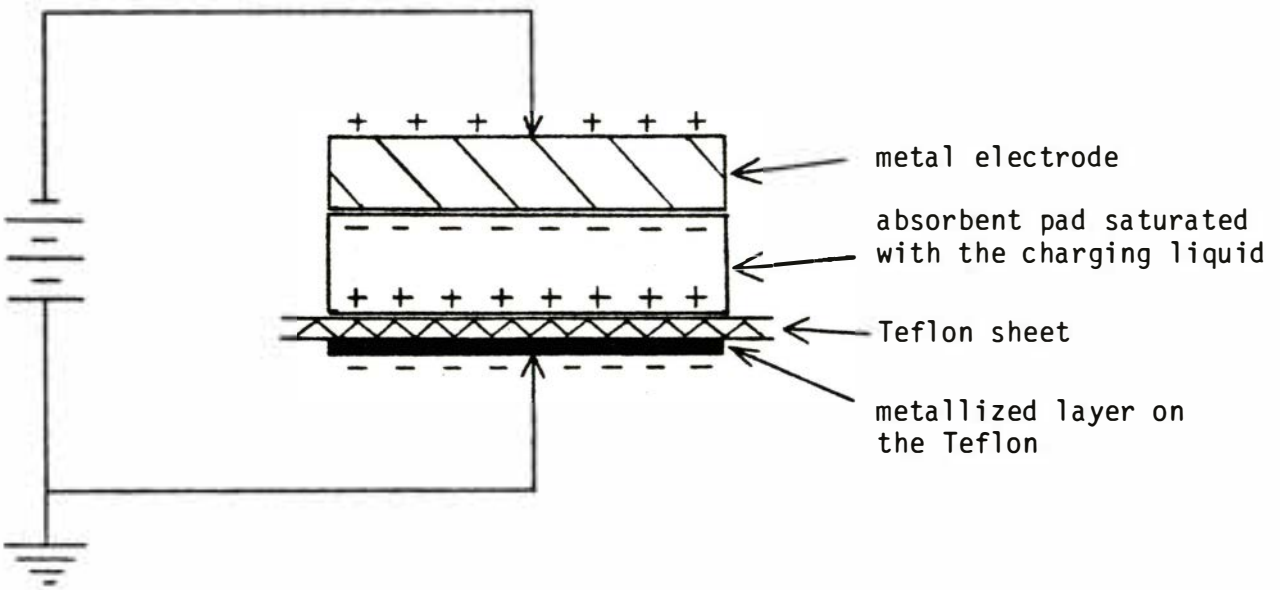

Fig. 1 A sketch of the electret production apparatus for the liquid contact formation method (not to scale).

and 0.31 cm-thick Teflon. Our charging times and voltages extended over a wide range of values at which useable electrets should have been formed.

For charging liquids, both ethanol and 0.01M NaOH solution were used. These were successfully tested by Chudleigh, while Kotrappa et al. used ethanol. Chudleigh also tested 0.01M HCℓ solution and water successfully, so it would seem that the actual liquid used is not a very critical parameter. The absorbent pad used to contain the charging liquid has been made of cotton (Chudleigh) and Whatman filter paper (Kotrappa et al.). We tested both materials, including finely woven cotton of thicknesses from ∿ 0.2 to 2 mm and cotton fiber of ∿ 4 mm.

Both methods of breaking contact between the charging pad (either connected or unconnected to the upper electrode) and the Teflon were tested (vertical and sliding movement). The overall size of the Teflon sheet containing the metallized area was also varied, in order to eliminate the possibility of surface conduction as a cause of failure. The original samples were 3.18 cm in diameter with a 2.54 cm metallized region. Samples up to 10.2 cm square were tested.

The resultant Teflon samples were all somewhat similar. A relatively small charge was collected on the Teflon. The charge was (+) or (-) as the upper electrode was (+) or (-). It was of a single polarity so that an electrometer registered about the same reading no matter which side of the Teflon was presented to it. The electric fields measured with the electrometer were about 10 to 100 volts/cm for a 1-cm air gap, when measured immediately after the charging period. A substantial fraction of the charge on the more highly charged samples leaked away within a few hours. The samples were therefore charged approximately as much as samples which are washed in water. There is a net absorption of (-) ions from the water during washing, as demonstrated by Chudleigh. This was confirmed by measuring washed samples with the electrometer, where they were found to have electric fields of about 10 to 20 volts/cm for a 1-cm air gap.

The reason for our inability to fabricate electrets by the liquid contact method could not be ascertained. It was decided to try the heated-sample method of production. The oven temperatures used by Kotrappa et al. (1980, 1981a) were reported to be 230°C and 240°C, with a 4.5 kV charging voltage and a 4-hour oven time, for 0.08 mm-thick samples. We used oven temperatures or 230°, 250° and 280°C for Teflon FEP and of 230°, 250°, 300° and 400°C for Teflon TFE for oven time of 4 hours. Charging voltages of 2000, 4500, and 6000 volts were tried. The results were similar to those obtained with the liquid contact method.

In none of the relevant literature is there any indication that special precautions or preparations are necessary in electret production. Annealing of the Teflon samples, before and after charging, may have some effect on stabilization of the charge. This is a secondary effect, however, and we could find no substantial annealing effect. Chudleigh (1976) mentions only that he used "virgin" material for his published measurements (that is, not previously used). Kotrappa et al. (1982) say the Teflon can be re-used at

a later date when the charge needs to be renewed.

In view of the difficulty in reproducing the published results in electret fabrication, this approach to radon daughter nuclei collection was set aside. If the problems can be resolved in the future, we may revert to this technique.

ELECTRICAL COLLECTION RDC DESIGN

The RDCs for electrical collection were designed to be tested with supplied DC power, as with batteries. Sketches of the relevant components are shown in Figure 2. A voltage is applied between the detector assembly and the Aℓ portion of the chamber walls. The 6 μm Aℓ overlayer has tags which loop over the CR-39 sample to contact the brass plate. The Aℓ overlayer is made (-) and collects on its surface the (+) charged radon daughters, which will be primarily RaA (^{218}Po). The Aℓ walls are made (+).

The volume of the chamber is 301 cm^3 and the dimensions are such that the walls are approximately equidistant from the detector, given the rectangular construction. For readout purposes it is desirable to have the track densities relatively equal across the surfaces of the CR-39 PNTDs. A hemispherical volume would be optimum in terms of equal electric field strengths throughout, which should translate into approximately equal densities of daughter nuclei collected over the negatively charged surface.

DETECTOR CALIBRATIONS

The RDCs were calibrated for collection potentials between 0 and 500 volts for relative humidities of 50% and 100%. The radon exposures were determined through direct comparison with calibrated RDCs of the smaller, non-electric type which were simultaneously exposed. Exposures were made in a laboratory radon exposure chamber. The CR-39 samples were removed from the exposed detectors at least 3 hours after the removal of the detectors from the exposure chamber to allow complete decay of the collected daughter nuclei. The CR-39 was etched in 6.25 NaOH solution at 70°C for 3.5 hours. Track densities were counted under a microscope at 200x. The track densities were found to be quite variable so that fields were counted in strips across the samples and average track densities were calculated. A counted strip is demonstrated in Figure 3. In addition to the variable track density it can be seen that the pattern of the tracks is somewhat off-center. This makes it difficult to standardize the counting of the samples.

When examined under reflected light the patterns formed by the etched tracks in the PNTDs could be clearly seen. Some of them are approximately reproduced in Figure 4. The track distributions are dependent on both collection voltage and relative humidity. None of the patterns are quite centered and the non-uniformity increases with voltage. The size of the areas with high track densities decreases as humidity increases.

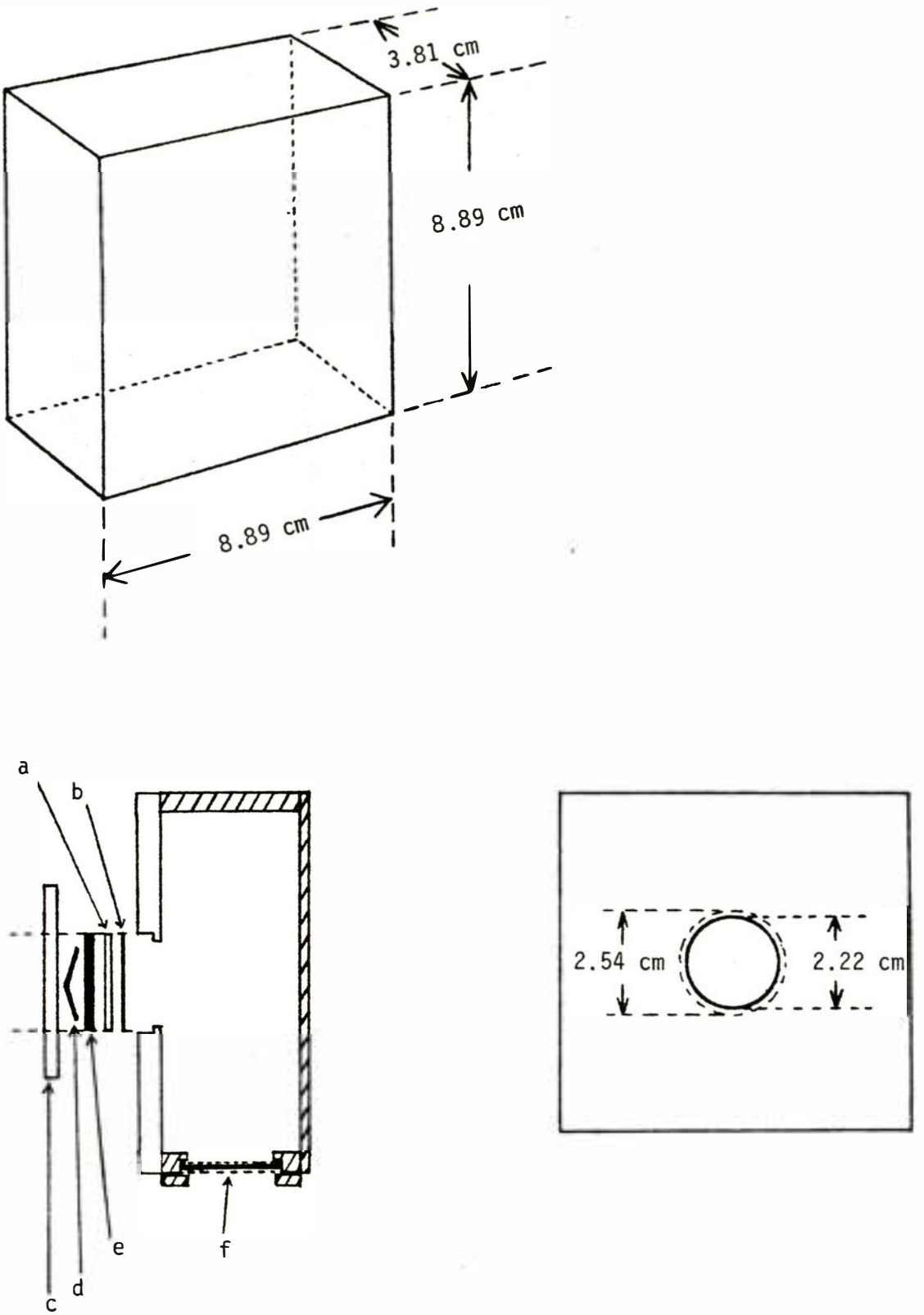

Fig. 2 Sketches of the electrical collection RDC for use with a battery. At top is the chamber volume. Below (left) is a side view showing the detector components: (a) CR-39 PNTD, (b) a 6 μm Aℓ overlayer, (c) acrylic cover plate, (d) steel spring band, (e) copper plate and (f) porous filter between two wire screens. The front and sides of the detector (hatched) are Aℓ and the back is acrylic. The total detector diameter and the diameter of the exposed area are shown at the right.

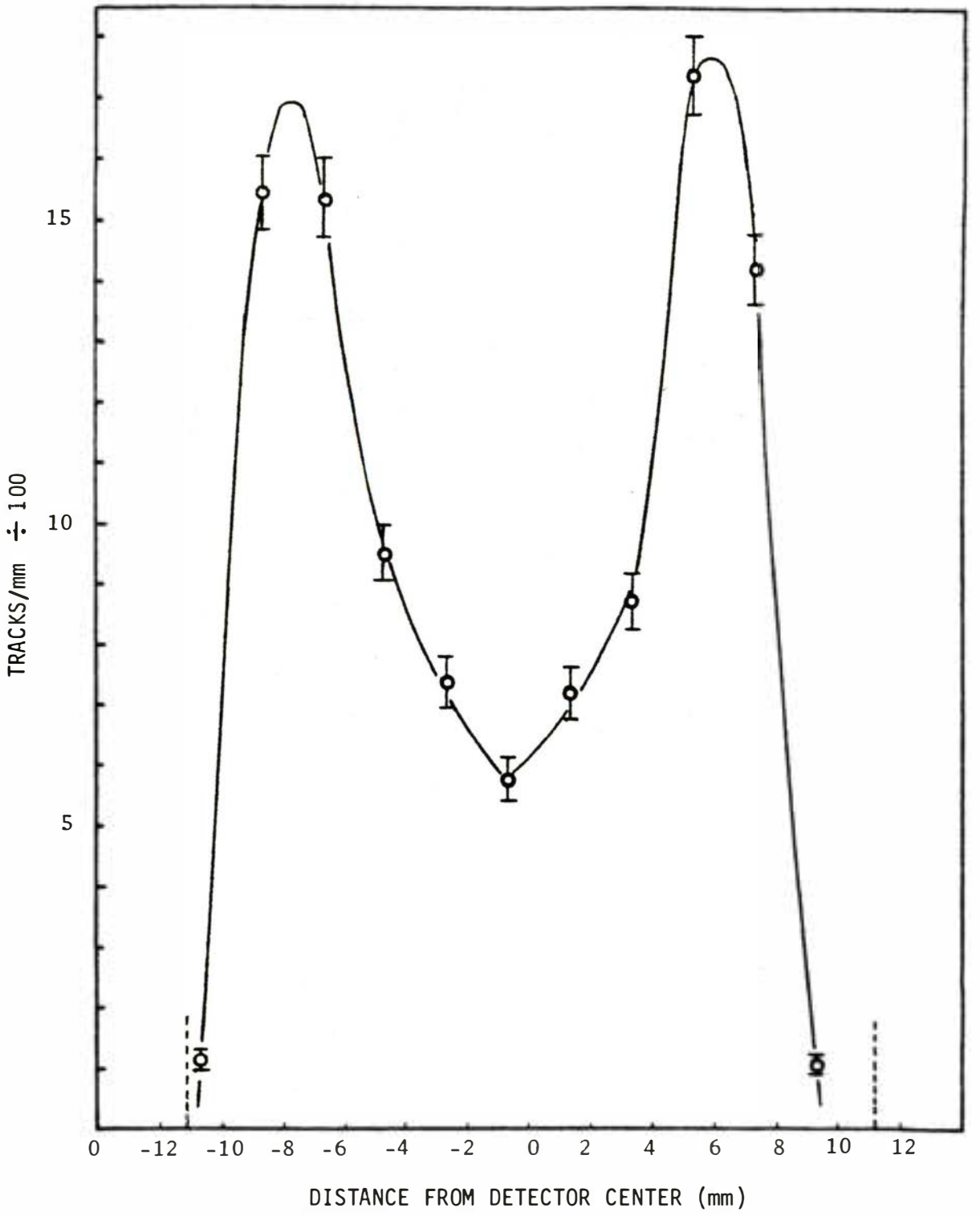

Fig. 3 Track density versus distance across the center of an exposed CR-39 PNTD. The dashed lines enclose the exposed portion of the sample. The collection voltage was 112 volts, relative humidity was 50% and the exposure was $\sim 6 \times 10^4$ pCi-hr/ℓ.

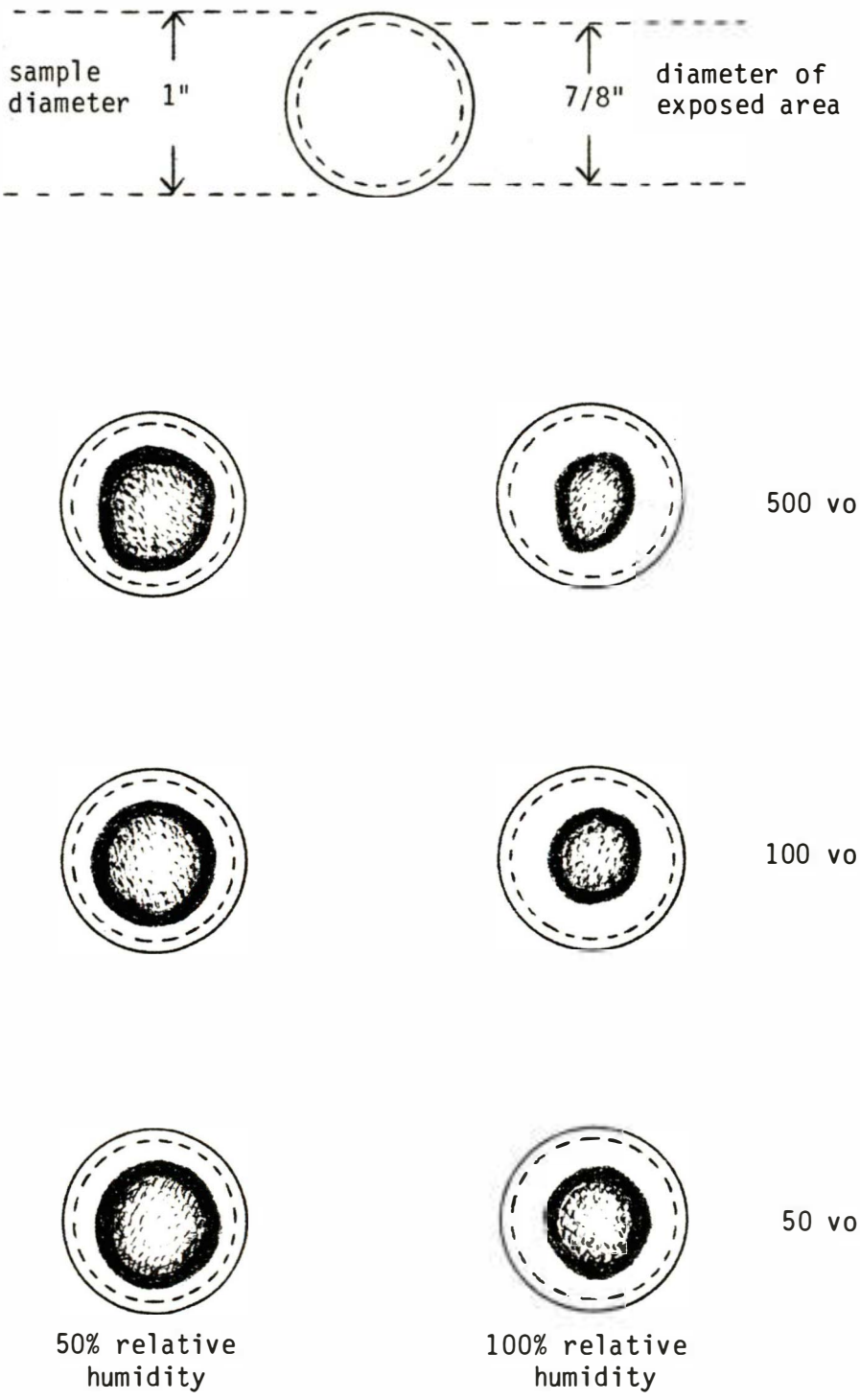

Fig. 4 Pencil reproductions of the patterns formed by alpha tracks on the exposed CR-39 PNTDs.

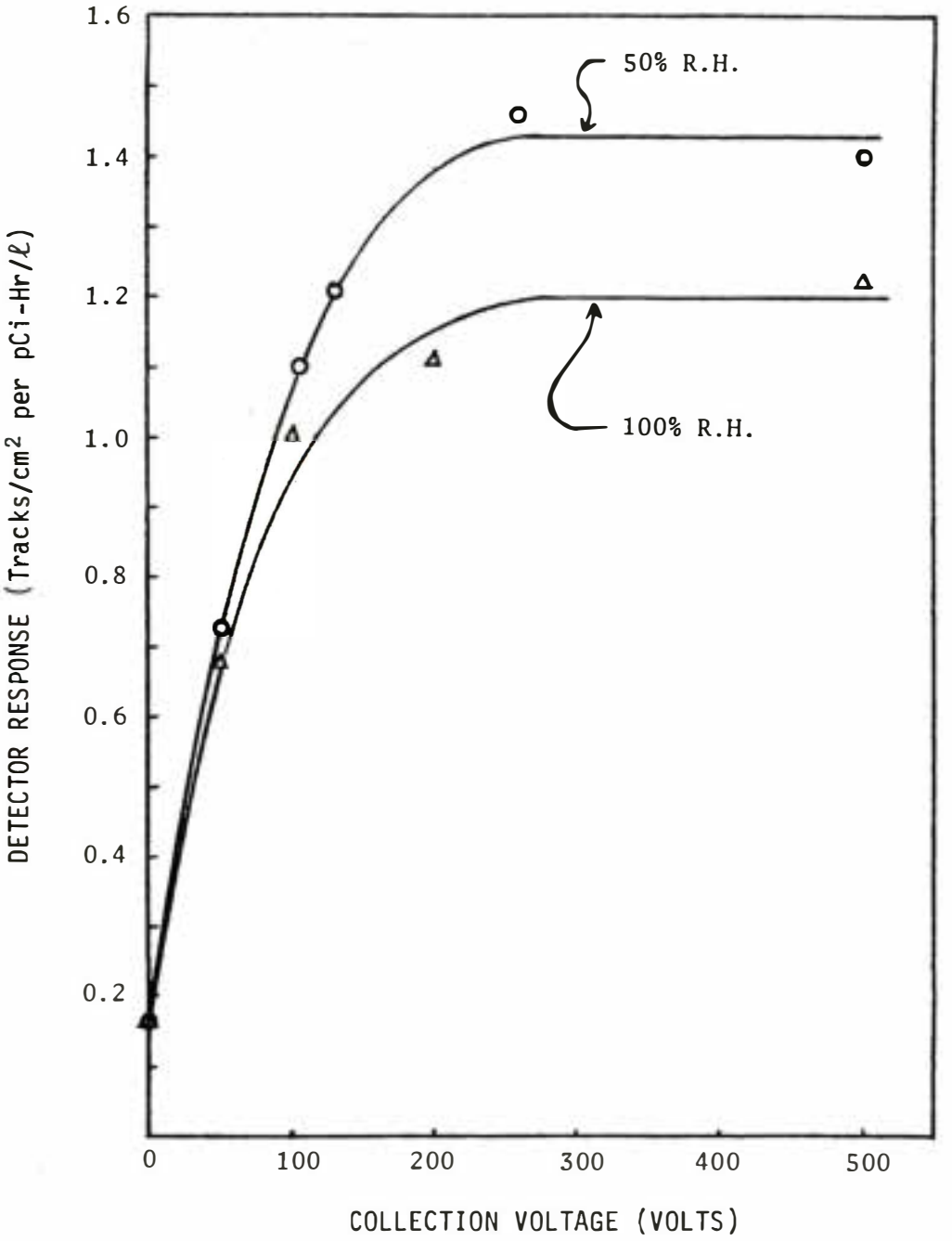

Fig. 5 Calibrations of the electrical collection RDCs versus collection voltage for 50% and 100% relative humidity.

The counted tracks were converted to an average track density over the 2.22 cm diameter exposed area of the PNTDs. The results are given in Figure 5. The deviations of the measured points about the fitted lines are mainly due to the non-uniformity of the tracks and the resulting counting errors. In the plateau region, for voltages above 250 volts, the response varies from 1.2 to 1.43 tracks/cm^2 per pCi-hr/ℓ. This is about 14 to 17 times greater than the responses of the more sensitive of the small non-electric RDCs used earlier. The volume of the electrical collection RDC is 40.7 times greater than that of the small RDCs so the difference in the responses is smaller than expected. With only 50% collection of the (-) radon daughters, one would still expect about 3 tracks/cm per pCi-hr/ℓ so it appears that the efficiency of collection is lower than expected. The exposed area of the PNTD is recessed behind an acrylic lip by 0.16 cm. Some of the daughters may have been lost by plate-out onto the acrylic.

Exposure times varied from 2.7 to 3.9 hours. If the radon level within the RDCs had not reached equilibrium with the surrounding air within a time short with respect to the exposure period (a few minutes), a loss of sensitivity would result. However, there was no correlation found between exposure time and sensitivity, which indicates that this was not a problem. The area of the porous filter on the RDCs was 13.2 cm^2, allowing a rapid diffusion of radon into the chamber.

SUMMARY OF MEASUREMENTS

The present design of the electrical collection RDC has the disadvantage of producing a quite variable distribution of track densities over the surfaces of the PNTDs. This makes the standardization of an accurate counting procedure difficult. Some redesign of the chamber would be necessary before routine use. The reasons for the observed ring of high track density, and the dependence of the diameter of the ring on humidity and collection voltage, are not understood at this time. It is likely that an electret rather than the battery-supplied voltage would produce a different pattern of track density since the electrical field lines within the chamber volume would be aligned differently. For a battery-supplied collection voltage, different chamber dimensions, in particular a lenthening of the chamber front-to-back and a reduction in the distances to the chamber sides, would be tested. The objective would be to flatten the track density distribution. An increase in efficiency of collection of the radon daughter nuclei would also be realized in this way by reducing end wall plate-out around the periphery of the PNTD.

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