IN-SITU PASSIVE MONITORING OF ALPHA-EMITTING RADIONUCLIDES

R. B. Gammage, J. C. DePriest, M. E. Murray
Oak Ridge National Laboratory*
P. O. Box 2008
Oak Ridge, Tennessee 37831-6383

R. V. Wheeler and M. R. Salasky
Landauer, Inc.
2 Science Drive
Glenwood, Illinois 60425-1586

J. C. Dempsey and P. Kotrappa
Rad Elec, Inc.
270 Technology Park
Frederick, Maryland 21701

*Managed by Martin Marietta Energy Systems, Inc., under contract number DE-AC05-84OR21400 for the United States Department of Energy.

IN-SITU PASSIVE MONITORING OF ALPHA-EMITTING RADIONUCLIDES

R. B. Gammage, J. C. DePriest, M. E. Murray
Oak Ridge National Laboratory
P. O. Box 2008
Oak Ridge, Tennessee 37831-6383

R. V. Wheeler and M. R. Salasky
Landauer, Inc.
2 Science Drive
Glenwood, Illinois 60425-1586

J. C. Dempsey and P. Kotrappa
Rad Elec, Inc.
270 Technology Park
Frederick, Maryland 21701

ABSTRACT

Electrets and alpha-track detectors (ATDs) show considerable promise for inexpensive passive monitoring of alpha contamination on man-made surfaces or in soil. At the stringent Department of Energy (DOE) limit of 100 dpm/100 cm^2, the electret voltage drops 10 V in about 4 hours; 10 V is readily quantifiable since any reading of electret voltage is accurate to ±1 V. An analogous signal-to-noise ratio for the ATDs is obtained after an exposure time of about 3 hours. The alpha-track registration efficiency for CR-39 type plastic is about 70% with the background track density averaging 13 tracks/cm^2. Measurements for intercomparison were performed with electrets, ATDs, and conventional survey meters on a contaminated vinyl floor and a concrete loading dock. Agreement between different types of detector readings was satisfactory. Surface soil measurements, using an exposure time of 1 day, can detect contamination of just a few pCi/g. Preliminary horizontal mapping was conducted within and at the boundary of a plutonium contaminated area at the DOE Nevada Test Site (NTS). The means of making vertical profiles of subsurface contamination are being explored. Some problems that have to be overcome involve interference from natural radon, variable soil moisture, preventing moisture condensation, wide extremes of ambient temperature and wind-driven shifting of soil.

INTRODUCTION

Contamination by alpha-emitting radionuclides is a pervasive and multi-faceted problem at most DOE weapons facilities. Alpha contamination is difficult to detect on narrow or irregularly shaped objects, in difficult to access or small spaces, and within soils. At the DOE limit of 100 dpm/100 cm^2, conventional hand-held survey meters have limited applicability.[1] Alternative methods of on-site detection are needed to meet some of the unfilled needs for characterization and decontamination and decommissioning, including verification of cleanup to very low levels. Recent publications highlighting two new approaches include an active long-range alpha surface monitor[2] and a passive alpha-track detector.[3]

In this ongoing study we evaluated the applicability of two types of modified, commercially available radon monitor to the passive monitoring of alpha-contaminated, man-made surfaces and soils. The findings are an expansion of studies reported at a recent symposium.[4]

METHODS

The ATDs were the CR-39 type of plastic with low background track density.[3] Exposures were made mostly with small chips, 1.9 cm^2 or 8 cm^2 in size. After exposure, the CR-39 chips were mailed to Landauer, Inc. for processing. Chemical etching of alpha damage tracks and subsequent automated track counts were completed to arrive at a track density from which the surface alpha activity was computed. The CR-39 does not respond to gamma or beta radiation.

The electret provided by Rad Elec, Inc., is a small ionization chamber, 3 cm in height, with an open face of 49 cm^2 area.[5] The alpha radiation emitted from a contamination area overlain by the chamber produces ionized air. The positive ions are drawn to and discharge a negatively charged Teflon plate; the exposure is computed in the field by making simple before-and-after voltage measurements using a small voltmeter. The sensitivity is strongly biased toward short-range alpha radiation should the electret encounter mixed alpha-gamma radiation fields. The response of the electret is little affected by changing atmospheric humidity, except in the extreme where water drops condense on the face of the electret.

Each type of device was calibrated with a ^{239}Pu or ^{241}Am check source of known activity in order to convert each field measurement on a man-made object into a contamination in dpm per unit area. Measurements indoors were made by placing the passive monitor directly in contact with the surface known to have fixed alpha contamination. In the case of soils brought to the laboratory, samples were weighed and then sprinkled onto a sticky pad. The passive monitors were then placed directly on top of the sticky pad-soil. In making direct soil measurements in the field, the devices were simply placed on the ground in a flattened footprint. The ATDs were weighted down with wood blocks to prevent them from blowing away.

RESULTS AND DISCUSSION

Calibrations were made with small ^{239}Pu and ^{241}Am alpha sources. The range of linearity of response of electrets extends from the fully charged condition, about 800 V, down to about 200 V of remaining charge. The response of an electret to ^{241}Am is shown in Figure 1. The "efficiency" factor for Am-241 is 7.7×10^{-4} ΔV/disintegration. At 100 dpm/100 cm^2, which is the most stringent DOE limit[6], a 10 V drop would occur in about a 260 minute exposure. The accuracy of an individual reading is ±1 V so that 10 V is a readily qualifiable signal. The drop in voltage during exposure on an uncontaminated surface is 3 V per day.

The alpha registration efficiency of the ATDs is about 70% while the background track density averages 13 tracks/cm^2. A signal-to-noise for the ATD, which is analogous to a 10 V voltage drop in the electret, requires an exposure time of about 3 hours.

The ATD provides a permanent record and a sub-mm spatial resolution of the alpha activity. Figure 2a shows the distribution of the alpha activity within a circle of approximately 2 cm dia. on a ^{241}Pu check source while Figure 2b reveals the distinctly different images of small "hot" particles on the surface of contaminated equipment. The "hot" particles produce spray-like clusters of tracks. The large numbers seen in Figure 2b are laser-etched markings used for identification. Each number is about 2 mm in height. The ATDs can distinguish between uniform and non-uniform alpha radiation fields as shown in Figure 3; a uranium metal source produces the homogeneous pattern of tracks in Figure 3a while the inhomogeneous track pattern of Figure 3b results from exposure of an ATD on a small metal flange attached to a system of contaminated piping.

An intercomparison was made of the capabilities of electrets, ATDs, and conventional hand-held survey probes. Two test locations where used, one indoors and one outdoors. To bring the areas surveyed by each type of device into rough correspondence, grids of 15 of the small ATD chips were employed that covered an area of 26cm^2. The ATDs and electrets (49cm^2) were each exposed for 20 minutes at both locations. The results of these field tests are listed in Table 1. The results are in reasonably good agreement with one another. The one mismatch, produced by the survey probe on the concrete, is explainable because of a marked localization of contamination within a surface pit in the concrete and the large area face (70cm^2) of the survey probe. The spatial distributions of the contamination, and the marked localization of contamination on the concrete, are revealed in Figure 4.

Table 1. Preliminary intercomparison of active and passive devices in monitoring fixed alpha contamination

Location	Survey probes (70 cm^2)	ATD (26 cm^2)	Electret (49 cm^2)
		-----dpm/100 cm^2-----	
Contaminated vinyl floor under glove box	3300-3800	3500	4300
Outdoor concrete loading dock	1200	2800	3000

The electrets and ATDs have also been evaluated for the more difficult application of monitoring soils contaminated with alpha emitters. Here we report on our attempts to make laboratory and field measurements on soils contaminated with plutonium. The soils tested were from, or at, the DOE NTS.

Three dry soil samples were evaluated in the laboratory at Oak Ridge National Laboratory. The nominal activities of the three soils were 0, 5, and 27 pCi/g. The "zero" activity sample was a supposedly uncontaminated soil taken from the desert adjacent to a car parking lot about 1 km from the contaminated zone. Aliquots of 2 g of fine soil particles were sprinkled evenly on sticky pads, which were cut to the same diameter as the diameter of the electret ion chamber.

The response factors per pCi/g for the electret and the ATD were 0.15 V/h and 9.6 tracks/mm^2-h, respectively. The electret, however, produced a large variability in response to different aliquots taken from the same bulk sample, as seen in Figure 5 for the 27 pCi/g soil. It remains to be determined whether this variability is caused by inhomogeneous distribution of the plutonium in the soil or to other factors. The number of ATD measurements to date are too few to comment on the precision. Our objective in this instance is to develop a simple and cost-effective screening test for measuring contamination in soil samples returned to a mobile laboratory and dried to a standard moisture content.

Preliminary in-situ tests were conducted on the desert floor at the DOE NTS. The testing occurred during a period of excessively hot weather accompanied by high winds that caused shifting of fine soil. Measurements were made at three different locations; at two points within an exclusion zone with plutonium surface contamination, at several points along the fenceline of the same exclusion zone and on the uncontaminated desert floor adjacent to the aforementioned parking lot. The "conversion" factors for bulk soil samples dried in the laboratory were used to convert the measurements on the desert floor to pCi/g equivalents; 0.256ΔV/h and 0.013 tracks/mm^2 per pCi/g. The results are contained in Table 2; there is reasonable agreement between soil activities measured by three different techniques, especially for a first scoping test where spatial inhomogeneities in contamination will affect each type of reading differently because different areas of soil are being interrogated. The potential usefulness of the ATDs for direct ground measurements is clearly evident, both in highly contaminated zones and in marginally contaminated areas only slightly above background levels. The possibility of interference by radon during longer exposures needs to be addressed in the future; differential measurements in pairs of detectors can be made by employing an alpha-particle absorber, which is permeable to radon, inside one of the pair of detectors. In future NTS visits, vertical profiling of contamination in the ground will also be addressed using strips of ATD material lowered into narrow-width holes made with a stake.

Table 2. Direct ground measurements with ATDs within or close to a plutonium contaminated area at the Nevada Test Site

		Equivalent (pCi/g)		
Location	Exposure Time	ATD	Electret	NaI*
Position close to ground zero	~ 10 min.	9800	4100	10,800
Position 23 m from ground zero	~ 10 min.	3300	2500	3500
Ten points along fence line at 10 m intervals (excess over car park below)	24 h	0-73	-	-
Uncontaminated desert floor adjacent to car parking lot	22 h	7	15	-

*Sodium iodide survey meter responding to 60 KeV X-rays from Am-241, which is the daughter of Pu-241.

CONCLUSIONS

Although the reported tests were intended to be only of a scoping nature, it is evident that both the electret and ATD are cost-effective, alpha-contamination monitors having promise in a variety of field screening applications. Both types of passive monitor have adequate sensitivity for making in-situ measurements on alpha-contaminated, man-made surfaces even at the most stringent release limits (100 dpm/100 cm^2). Intercomparison testing of passive monitors with conventional survey meters has already produced satisfactory results for outdoor concrete and indoor vinyl floor surfaces. The electret's principal advantage is the ability to make direct screening measurements in the field using an accompanying, hand-held voltage meter to make before-and-after exposure readings of the decreasing electret voltage. The attractiveness of the ATD is that it can be precut to convenient size, it has the ability to reach small and difficult to access locations, it can spatially resolve the contamination, identification of "hot" particles is possible and a permanent recording of the radiation field is obtained.

The passive monitors can also be used to screen contaminated soils. The detectors have monitored soil containing only a few pCi/g of plutonium using exposure times of 24 hours. Further testing is needed to resolve problems of interference by radon, possible adverse response under high or low ambient temperatures, rain or snow, and soil shifting in high winds. During the next field trip we will attempt vertical profiling of soil contamination.

REFERENCES

1. J.B. Martin, D.M. Fleming, K.L. Swinth, Practical Limitations on Alpha Contamination Monitoring, Abstract, THPM-B4, S52, Thirty Fifth Annual Meeting, Health Physics Society, 1990.

2. D.W. McArthur, K.S. Allander, K.B. Butterfield, "Long-Range Alpha Detector," Health Phys. 63:324-330 (1992).

3. R.B. Gammage, R.V. Wheeler, "Preliminary Testing of CR-39 for Surface Alpha Contamination Monitoring," Health Phys., in press.

4. R.B. Gammage, J.C. DePriest, R.V. Wheeler et al., "Monitoring of Surface Alpha Contamination with Inexpensive Passive Monitors," in Proceedings of the 1992 International Symposium on Environmental Contamination in Central and Eastern Europe, Budapest, Hungary, October 12-16, 1992, in press.

5. P. Kotrappa, J.C. Dempsey, R.W. Ramsey, L.R. Stieff, "A Practical Electret Passive Environmental Radon Monitor for Indoor Radon Measurement," Health Phys. 58:461 (1990).

6. U.S. Department of Energy, Guidelines for Residential Radioactivity at Formerly Utilized Sites Remedial Action Program and Remote Surplus Facilities, Management Programs Sites (Revision 2, March 1987); ibid, DOE Order 5400.5 (April 1990).

KEY WORDS:

Alpha contamination
Passive monitoring
Electret
Alpha track detector
Plutonium
Surfaces
Soils
Hot particles

Figure 1. Rate of electret voltage drop during exposure to a ^{241}Am check source.

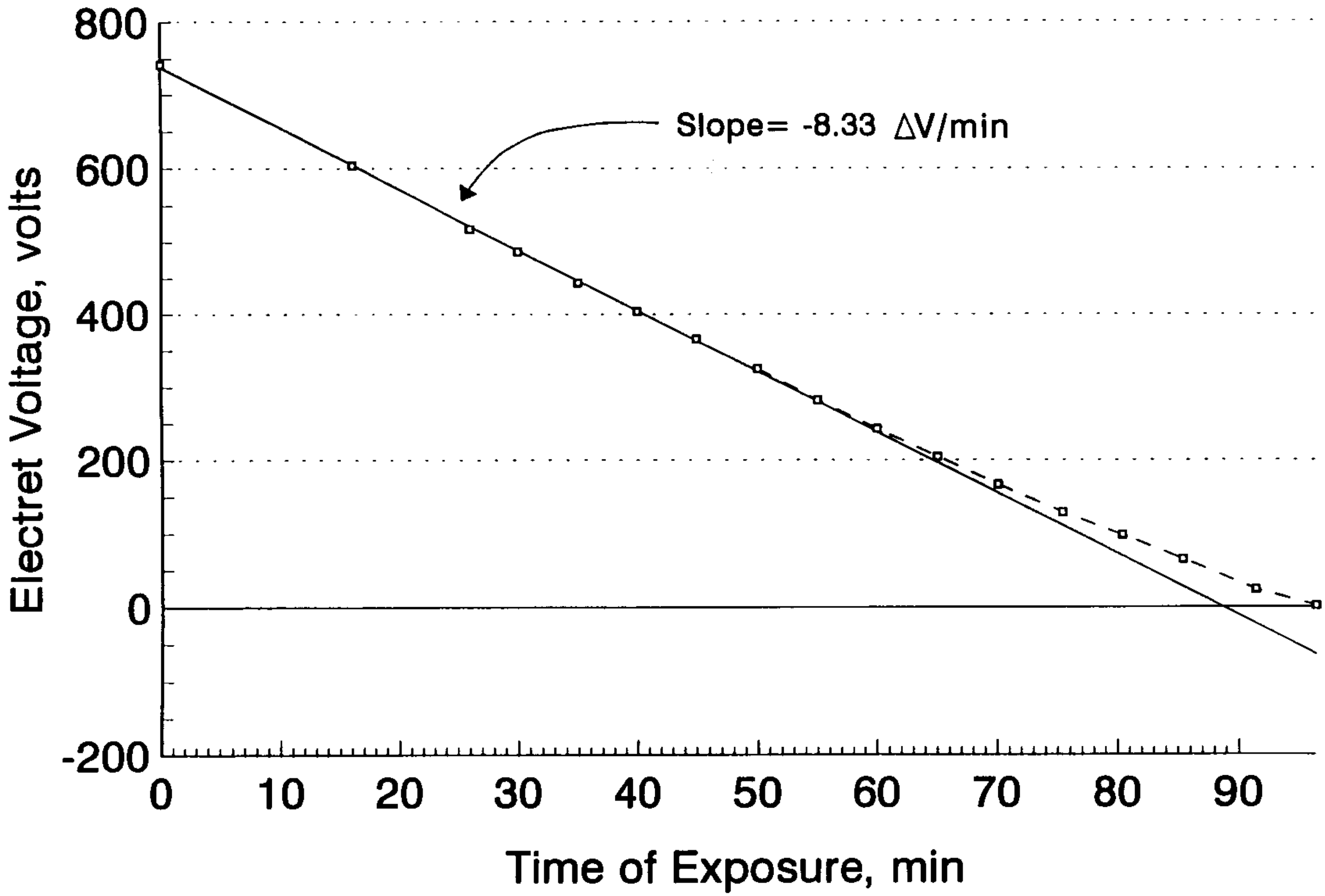
Figure 2. Photographs of etch pit patterns from (a) a Pu-239 check source (4.1cm x 4.1 cm square CR-39 plastic) and (b) a surface contaminated with particulate alpha emitters.

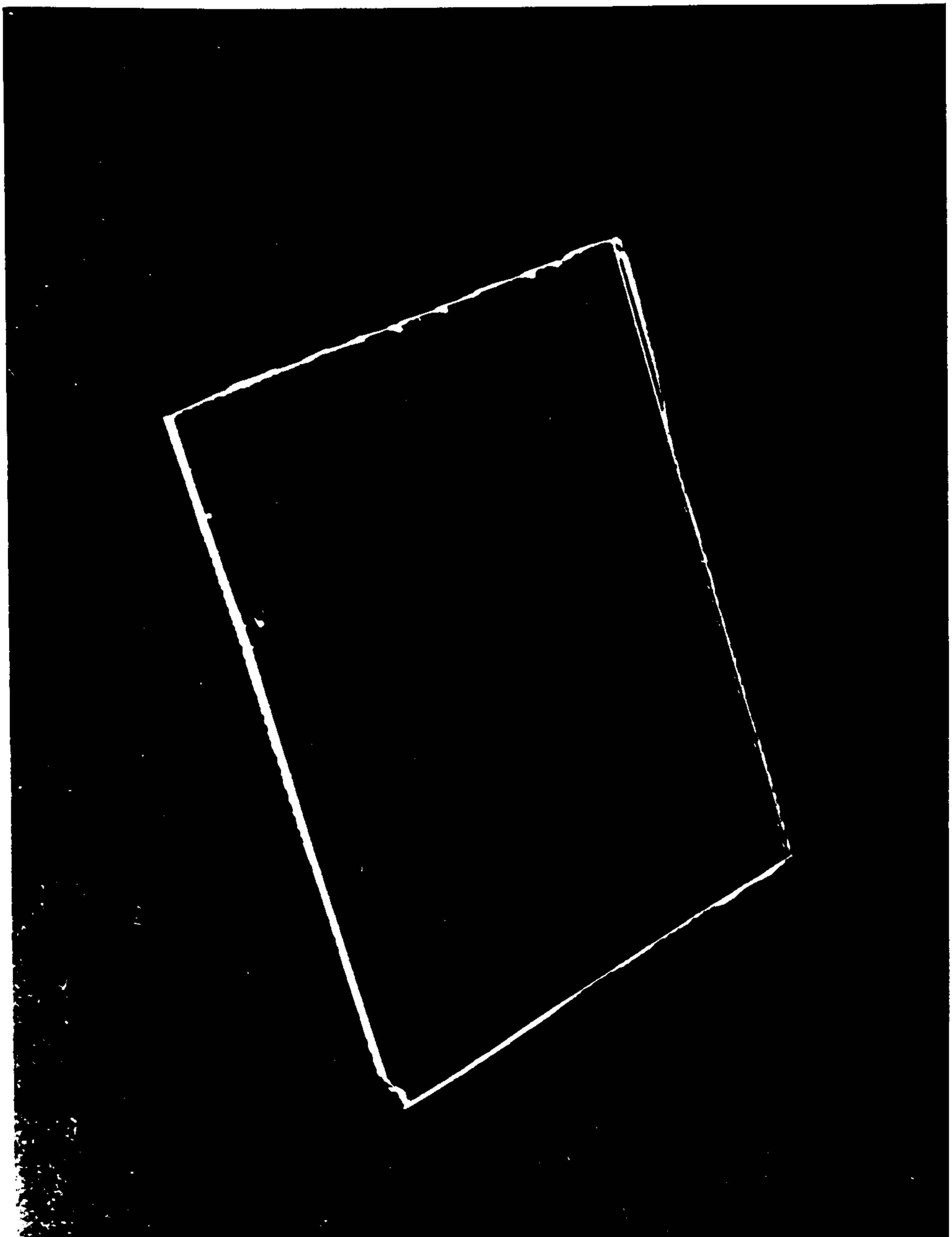
Figure 3. Photograph of etch pit patterns from (a) uranium metal and (b) an inhomogeneously contaminated valve flange.

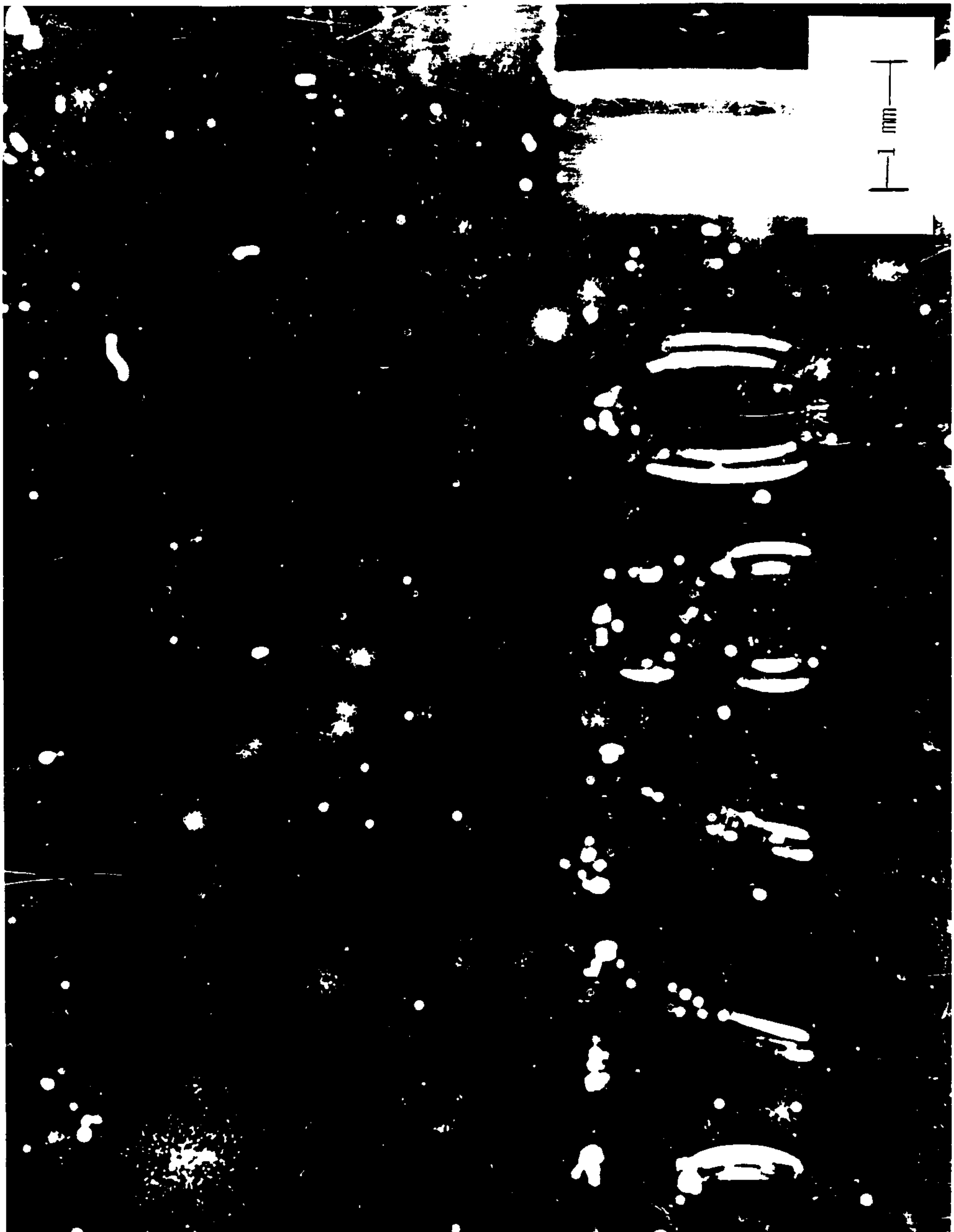
Figure 4. Maps of inhomogeneously distributed contamination (dpm/cm^2) measured by 15 chip grids of ATDs.

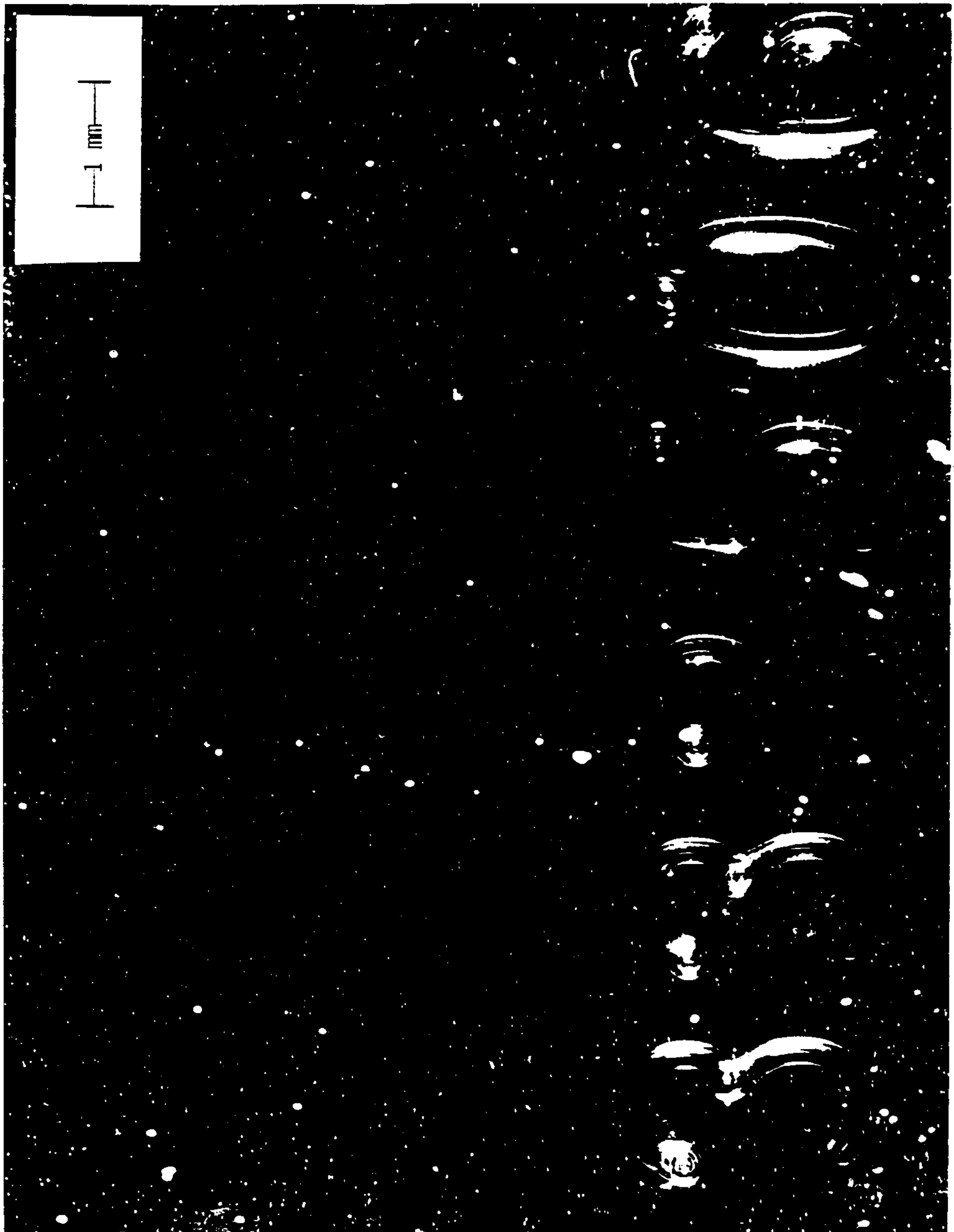
Figure 5. Voltage drop from four 2 g aliquots of fines from the same bulk soil sample.

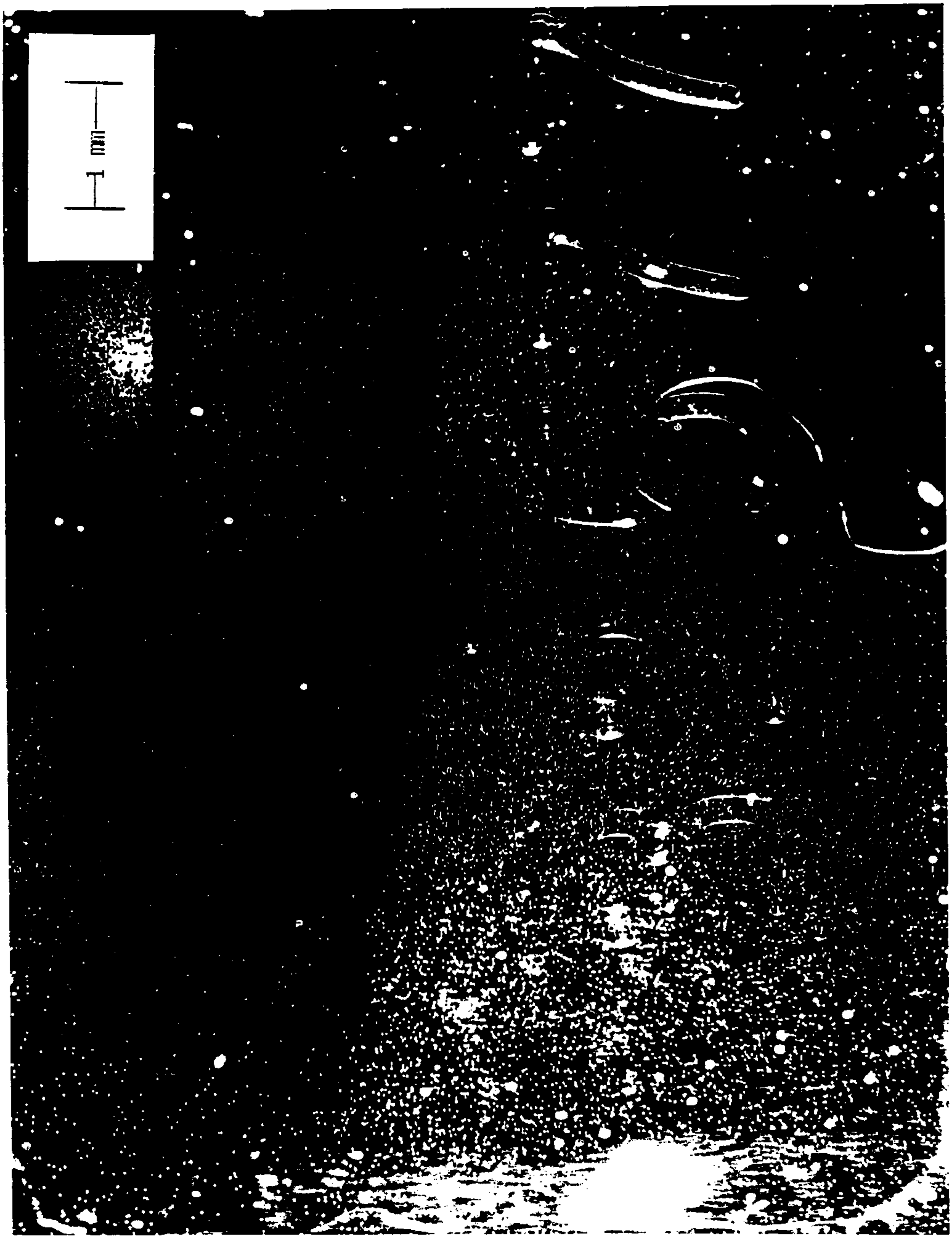
^{241}Am 10800 dpm

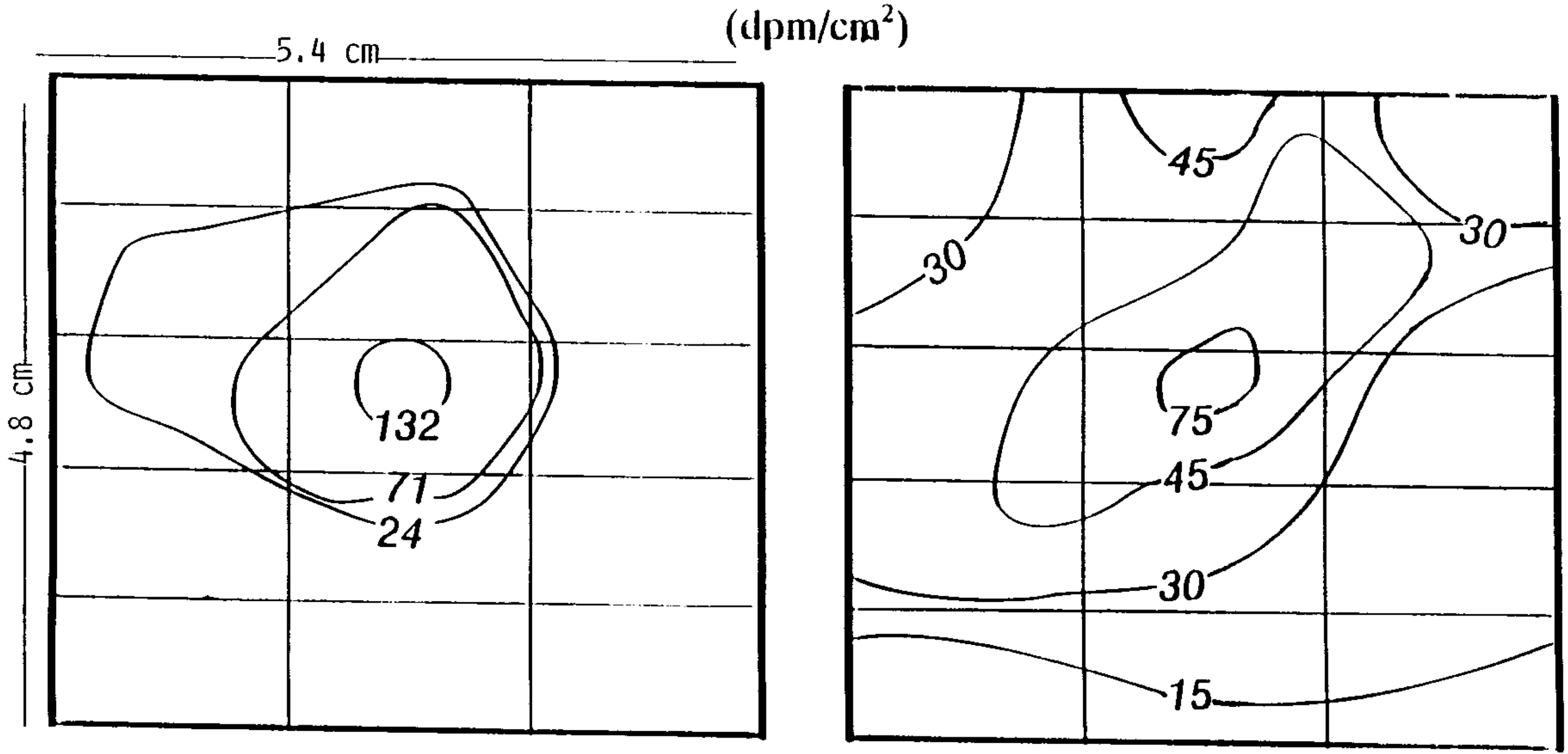





1 mm

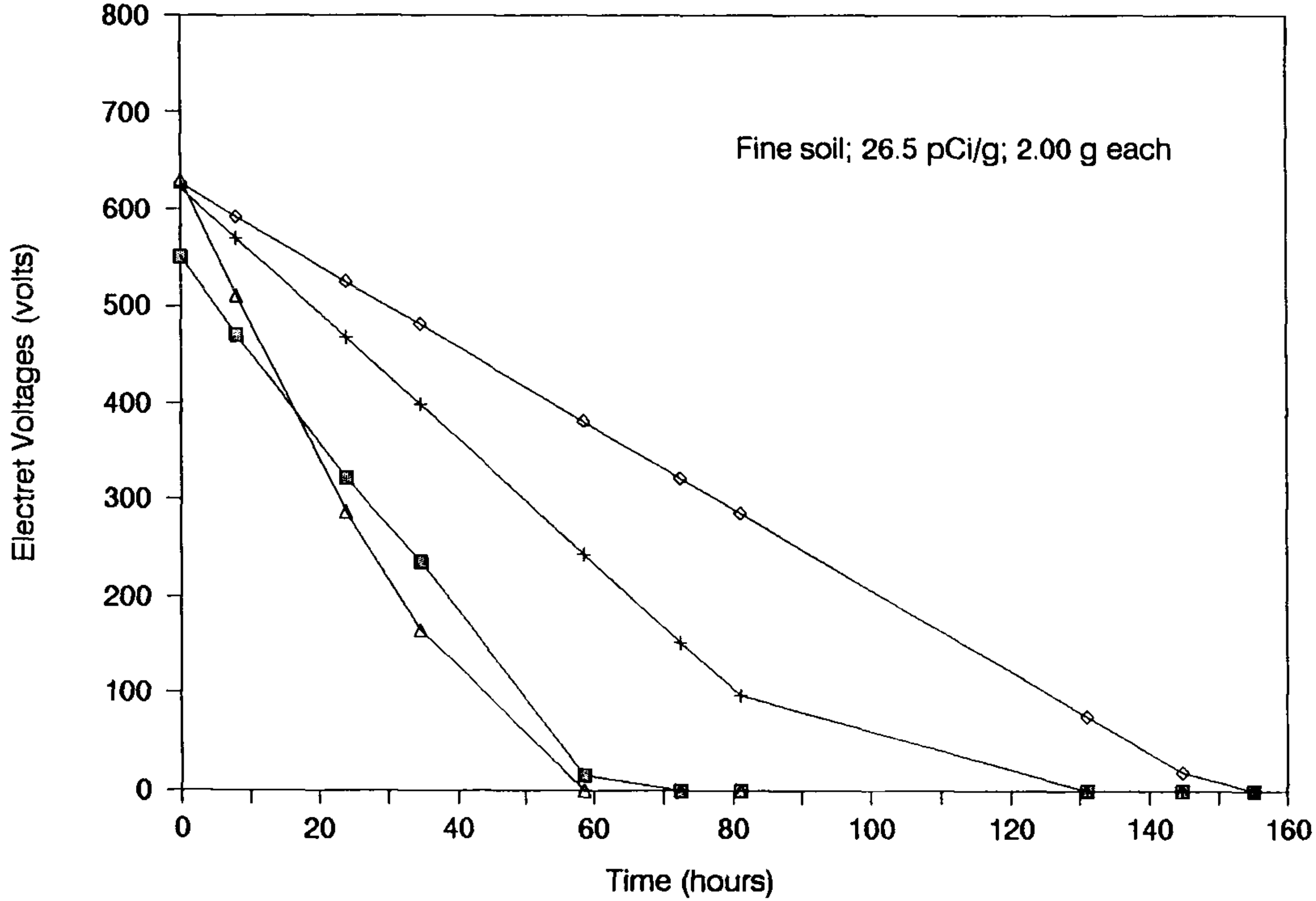

1 mm


1 mm



Outdoor Concrete Pad

Indoor Vinyl Floor



Fine soil; 26.5 pCi/g; 2.00 g each
Electret Voltages (volts)
Time (hours)
0
100
200
300
400
500
600
700
800
0
20
40
60
80
100
120
140
160