

ANL/RE/RP--87642

STATE OF THE ART OF D & D INSTRUMENTATION TECHNOLOGY
--ALPHA COUNTING IN THE PRESENCE OF HIGH BACKGROUND

by

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August, 1995

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List of Acronyms and Abbreviations

ALARA	As low as reasonably achievable.
ALI	Annual limit on intake. One ALI corresponds to a total 50-yr dose to the individual of 5 rem.
ANL	Argonne National Laboratory, Argonne, IL.
Bq	One disintegration/second.
Ci, curie	3.7×10^{10} disintegrations/second.
D&D	Decontamination and decommissioning.
DAC	Derived air concentration, defined as the ALI divided by $2,400 \text{ m}^3$. This corresponds to an individual receiving one ALI in an exposure time of 50, 40-hour weeks.
dpm	Disintegrations per minute.
Eberline	Eberline Instruments, Santa Fe, NM.
LANL	Los Alamos National Laboratory, Los Alamos, NM.
LC	Lucas Cell, a scintillation device for collecting and measuring airborne alpha activity.
LIF	Laser-induced Fluorescence.
LRAD	Long range alpha detector, technology developed by LANL for detection of ionized air generated by alpha particles.
MeV	Million electron-volts, a commonly used measure for the energy of alpha, beta, and gamma rays.
nm	Nano-meters, a commonly used measure for light wavelengths.
NORM	Naturally occurring radioactive material.
R/hr	Dose rate in rem/hour.
SSM	Soil-surface monitor.
STL	EG&G/Special Technologies Laboratory, Santa Barbara, CA.
TRU	Transuranic elements.
TRU waste	Waste contaminated by TRU at or above the level of 100 nanocuries/gram.



STATE OF THE ART OF D & D INSTRUMENTATION TECHNOLOGY

--ALPHA COUNTING IN THE PRESENCE OF HIGH BACKGROUND

C. E. Dickerman

Abstract

Discrimination of alpha activity in the presence of a high radiation background has been identified as an area of concern to be studied for D&D applications. Upon evaluating the range of alpha detection needs for D&D operations, we have expanded this study to address the operational concern of greatly expediting alpha counting of rough surfaces and rubble. Note that the term, "rough surfaces" includes a wide range of practical cases, including contaminated equipment and work surfaces. We have developed provisional applications requirements for instrumentation of this type; and we also have generated the scope of a program of instrument evaluation and testing, with emphasis on practical implementation. In order to obtain the full operational benefit of alpha discrimination in the presence of strong beta-gamma radiation background, the detection system must be capable of some form of remote or semi-remote operation in order to reduce operator exposure. We have identified a highly promising technique, the long-range alpha detector (LRAD), for alpha discrimination in the presence of high radiation background. This technique operates upon the principle of transporting alpha-ionized air to an ionization detector. A transport time within a few seconds is adequate. Neither the provisional requirements nor the evaluation and testing scope were expressly tailored to force the selection of a LRAD technology, and they could be used as a basis for studies of other promising technologies. However, a technology that remotely detects alpha-ionized air (e. g., LRAD) is a natural fit to the key requirements of rejection of high background at the survey location and operator protection. Also, LRAD appears to be valuable for D&D applications as a means of greatly expediting surface alpha-activity surveys that otherwise would require performing time-consuming scans over surfaces of interest with alpha detector probes, and even more labor-intensive surface wipe surveys. In addition, a true remote-viewing technique, laser-induced fluorescence (LIF), has been identified. Because of the limited information available, the suitability of laser-induced fluorescence for practical application to D&D operations is not clear at this time.

1.0 INTRODUCTION

Alpha-emitting radioactive contamination presents a special set of characteristics from the standpoint of decontamination and decommissioning (D&D) activities. Detection of alpha activity is important to protect workers, to help characterize contamination by actinide or transuranics (TRU), and to determine if specific items can be given unrestricted release from a radiological control area. Alpha-particles of a given energy are much more highly ionizing than beta-rays or gamma-rays. Alpha-radiation is readily absorbed, by skin, a few sheets of paper, a relatively thin layer of air (APPENDIX A.1), etc. Because alphas are readily absorbed by matter, the detection of alpha-radioactivity in practical D&D applications can be difficult and time-consuming. For example, a coating of paint could be sufficient to prevent detection of high alpha-radioactive contamination of a wall or floor. Alpha-radioactive contamination is an important concern, because the biological damage from an internal source of alpha-radiation, such as that inhaled by a D&D worker, is much more concentrated than that from beta- or gamma-emitters.

A specific concern identified for study in the D&D Technology program at ANL is the detection, or discrimination, of alpha-activity in the presence of high beta-gamma radiation background. It is particularly important for this discrimination to be achieved with a minimum of exposure to personnel performing the alpha surveys. Alpha contamination from uranium and other actinides is expected from uranium enrichment plants and reactor fuel manufacturing plants. Alpha contamination also is potentially present wherever spent reactor fuel has been stored. In addition, alpha-emitting naturally occurring radioactive material (NORM) occurs in any facility with concrete or masonry construction, as well as in facilities that handle or process naturally occurring radioactive material materials (oil and natural gas operations, rock phosphate fertilizer, ceramic mantles for gaslights, uranium mining, etc.). This naturally occurring radioactive material will need to be characterized to determine if it qualifies for release as normal industrial waste.

Two types of alpha-ray contamination detection cases are considered:

1. Surface alpha contamination usually is the most important alpha-detection application for D&D activities. Because alphas are absorbed in thin layers of material, bulk contamination gives rise to a detectable alpha flux only from a thin surface layer.
2. Airborne alpha-contaminated particulates also can be a significant hazard. These can arise from abrasion of contaminated material (D&D cutting operations, traffic through facility, etc.). Alpha-contaminated particulates also can arise from contamination of dust by the daughters of radioactive radon (Rn) gas. Radon occurs naturally, has atomic number 86, and exists in the form of several isotopes. Rn-219 has a half-life of 3.9 s, is both an alpha-emitter and a gamma-emitter, and is a daughter of the naturally occurring decay chain beginning with U-235. Rn-220 has a half-life of about 55 s, is an alpha-emitter, and is a daughter of the naturally occurring thorium radioactive chain beginning with Th-232. Rn-222 has a half-life of 3.8 days, is an alpha-emitter, and is a daughter of the naturally occurring radium isotope, Ra-



226. Rn isotopes also may occur as daughters of artificially produced transuranics. For example, one plutonium isotope (Pu-239) feeds the decay chain that produces Rn-219. (Also, there is one artificially produced radioactive chain, the neptunium chain, which produces a radon isotope, Rn-216. Rn-216 has such a very short half-life that its gaseous state may be ignored: it may be considered to be merely a short-lived alpha-emitting daughter of the short-lived Ra-220.)

Radon is a noble gas. It often is detected by collecting radon daughter-contaminated particulates on a filter and detecting radiation from the filter. Rn-222, with a 3.8 day half-life, is normally considered to be the dominant concern where radon gas is a problem.

We will not discuss survey of liquids for alpha contamination. Equipment for laboratory analysis of liquids for alpha contamination is available commercially. Laboratory processing of samples for alpha spectrometry (for example, to identify specific contaminants) performed to provide thin deposits on planchettes or dissolve samples in liquid scintillators will not be discussed here, either.

Our evaluation has identified another practical D&D concern that should be addressed also. It is important to be able to survey quickly and efficiently real working surfaces and real equipment, which rarely consist of flat, plane surfaces. It is also important that D&D operations be able to distinguish quickly and efficiently when surface removal of concrete and other contaminated structure has proceeded sufficiently that the remaining material no longer need to be handled as contaminated waste. As will be discussed briefly in Section 2, below, it is apparent that there are *significant and obvious problems in being able to reliably position a conventional alpha probe close enough to the surface to intercept a fraction of alphas sufficient to obtain good survey results for rough surfaces, and particularly for material such as rubble, etc.* Therefore, we have expanded the scope of this study to include the survey of rough surfaces and rubble for alpha contamination. Note that the term, "rough surfaces" includes a wide range of practical cases, including contaminated equipment and work surfaces.

Health physics surveys to determine the magnitude of activity present will need to make extensive use of slow and time-consuming smear surveys. If the details of the facility and facility equipment are such that alpha detector probes can be used, these probes still will have to be positioned close to the surfaces, and moved carefully and slowly to assure adequate coverage. More than one survey typically will be necessary for a specific portion of a given facility: There will need to be an initial survey to determine the extent of the radiological hazard present. Additional comprehensive surveys will be necessary to monitor progress in decontamination, and a final survey will be needed to determine that the work has reached the required level of decontamination.



2.0 TECHNOLOGY

Conventional alpha-detection instrumentation for health physics applications is a mature technology, which will be discussed briefly below as an introduction before proceeding to discussion of more advanced (exotic?) approaches.

2.1 Conventional Health Physics Instrumentation--Surface Alpha Activity

There is a wide range of types, sizes and shapes of portable, hand-held, battery-powered alpha detectors for health physics applications available from commercial sources. Commercial alpha detectors include gas detectors and scintillation detectors. Typically, these detectors are capable of high sensitivity, in order to be used for surveying down to the very low alpha-radiation levels required to qualify persons and items as "uncontaminated". Single-purpose alpha probes are available. Also, there are general-purpose survey meters designed to accept alpha sensitive probes. Because of the short alpha-ray range in air, these probes must be positioned close to the surfaces being surveyed, say, within a few millimeters. These probes may be used to survey surfaces directly, or they may be used to survey a piece of filter paper or other flexible material used to perform a surface smear survey on the object of interest. Typically, surface activity is reported in units of disintegrations per minute/100 cm² (dpm/100 cm²).

Gas detectors typically have thin windows (there are windowless detectors, also) to allow alpha particles to enter the sensitive gas volume. Windows consisting of aluminized mylar, with thicknesses of the order of one mg/cm² are typical. Because of the fragile nature of the window, protective mesh or grids may be placed over the window. Large-area scintillator alpha probes also are available commercially. These may have fine particles of ZnS scintillator embedded in the surface of a plastic, covered with a thin light-protective aluminum film. Scintillator probes are available with surface areas up to about 600 cm². Because of need to position the surface of the probe within a cm of the surface being surveyed, damage to windows and protective films is a continuing operational concern, particularly for surveys of rough surfaces and rubble.

Both gas detectors and scintillation detectors respond to ionization from beta-rays, as well as from alpha-rays, but alpha-ray pulses can be discriminated from beta-ray pulses by electronic circuitry. Gamma-ray sensitivity is low for alpha detectors of both types. Because of alpha energy losses in air, in detector windows, and in window mesh/grid structure, the energy spectra of alphas entering the sensitive regions of these probes are degraded from the original spectra. Thus, these probes are not sold as true energy spectrometers with practical ability to distinguish between alpha energy spectra from different isotopes.

Solid-state semi-conductor alpha detector systems are commercially available for use under laboratory conditions to obtain well-resolved alpha energy spectra that are good enough for identification of specific alpha-emitting isotopes. In some cases, proportional counters also are used for laboratory analyses.

Because of the need to position an alpha probe accurately, and the need to cover completely the surface areas of interest with a scanning speed slow enough to detect relatively small localized contamination, alpha probe surveys are time consuming. It is widely recognized that the process of performing surface smear surveys is even more time-consuming. (ANL, 1991)

2.2 Conventional Health Physics Instrumentation--Airborne Alpha Activity

Conventional health physics instrumentation for measuring airborne alpha activity provides a means of measuring alpha activity in locations where there is high beta-gamma background.

Alpha detectors are commercially available for monitoring airborne alpha contamination, as deposited on a filter. Typical detectors have much higher sensitivity for the highly ionizing alphas than betas or gammas; further, alpha pulses can be distinguished from beta and gamma pulses by electronic circuitry. These filters are designed to provide a well-defined geometry suitable for alpha energy spectrometry. The three principal radon daughter products decay with emission of alphas with relatively high energies: 7.5 MeV for the daughter of Rn-219, 6.9 MeV for the daughter of Rn-220, and 6.1 MeV for the daughter of Rn-222. Because the maximum alpha energy for alphas emitted by the plutonium isotopes Pu-239 through Pu-242 is only 5.15 MeV (and alphas from Pu-238 are 5.5 MeV), air monitors equipped with gas proportional counters or solid-state semi-conductor detectors can distinguish between airborne contamination from plutonium and contamination from radon + radon daughters.

Another type of device for measuring airborne alpha activity is the well-known Lucas Cell (LC). A typical Lucas Cell is a vacuum-tight cup-shaped chamber, of about 100 cm³ volume, closed with a transparent window and fitted with an evacuation valve. The inner surface of the cup is covered with a thin coating of ZnS(Ag) alpha-scintillator material. For use, the Lucas Cell is evacuated through the valve, which then is closed. Next, the Lucas Cell is taken to the location to be sampled, the valve is opened to admit the gas sample, and the valve is closed. The cell is then removed to the laboratory, attached optically to a photomultiplier tube, and alpha-induced scintillation events are counted, in a region remote from the survey location background. Semkow, et al., 1994, describe a state-of-the-art Lucas Cell.

2.3 Detection of Ionized Air

2.3.1 Long-Range Alpha Detector

One of the early techniques for the detection of ionizing radiation from radioactive sources was the electrometer, which detected the presence of ionized air between charged electrodes. One particularly simple form, of historical importance, used two thin foils as the electrodes: the foil separation was noted when the two foils were fully charged with the same polarity, and ionization was detected by observing the rate with which the two foils came together as charge was lost through the air. This type of instrument was used to detect the presence of ionizing radiation *at the location of the electrometer*. However, this ionization



typically has a finite lifetime, and it is possible to transport the ionized air from its volume of origin to an ionization detector *located at a distance from the radioactive source*. The remote detection of ionized air cannot discriminate between ionization produced by alphas, betas, and gammas. However, the very short range of alphas in air means that the intrinsic efficiency of an air-ionization detector is much higher for alphas than for the other forms of ionizing radiation. Five MeV is a typical alpha-ray energy for transuranics. Using a typical value of 35 eV energy loss per ion pair generated in air (Knoll, 1989), this alpha will generate about 150,000 ion pairs in a track length of about 3.5 cm. This is a very high ion concentration compared with those generated by a 5 MeV gamma ray or even a 5 MeV beta ray.

MacArthur and collaborators at Los Alamos National Laboratory (MacArthur, et al., 1992a) have developed a series of long-range alpha detector (LRAD) instruments that detect alpha-rays indirectly by collecting and measuring the electric charge on ions generated in air by the passage of alpha-rays. The developers consistently refer to this technique using the acronym, LRAD, and we will use LRAD throughout this report in order to avoid possible confusion with their literature. They have shown that it is practical to transport this ionized air to a detector located at distances of the order of several meters using a small fan.

Figure 2-1 illustrates the basic LRAD principle. The concept may be described as follows: a collection chamber is placed over the surface area to be surveyed. Ionized air produced by alpha particles in a relatively thin air layer above the surface is moved through a transfer tube by a fan into the sensitive volume of a charge detector. The output of the charge detector can be calibrated to yield the rate with which air is ionized by the radioactive material. Given the ionization rate and the dimensions of the collector, the dose rate can be calculated. The ionization rate also can be used to calculate back and estimate the alpha surface activity in disintegrations per minute per 100 cm². A wide range of LRAD designs may be developed, based on this general approach.

MacArthur, et al., (1992b) have reported results with a LRAD configuration with a moveable source located inside a 2.4 meter-long aluminum pipe having an inner diameter of only 3.4 cm attached to the front end of an ion detector, and an air velocity of about 16 meters/min (about 2.5 s for ions to travel the length of the pipe). The ion detector output with the source located 2.4 meters from the detector was about 70% of the signal detected with the source located at the entrance to the ion detector. This measurement means that the ionized air had a half-life of about 5 s when it was transported at this particular velocity in this particular geometry, a very encouraging result. The developers also have tested the instrument sensitivity to environmental effects including variations in humidity, temperature, barometric pressure, and airborne dust concentration. (MacArthur, et al., 1992a) It should be noted that the ion detector can be configured so that it simultaneously measures the radiation ionization background *at the location of the ion detector*, allowing automatic subtraction of this background from the LRAD signal generated by ionized air drawn in from the collector. (Johnson, et al., 1992).

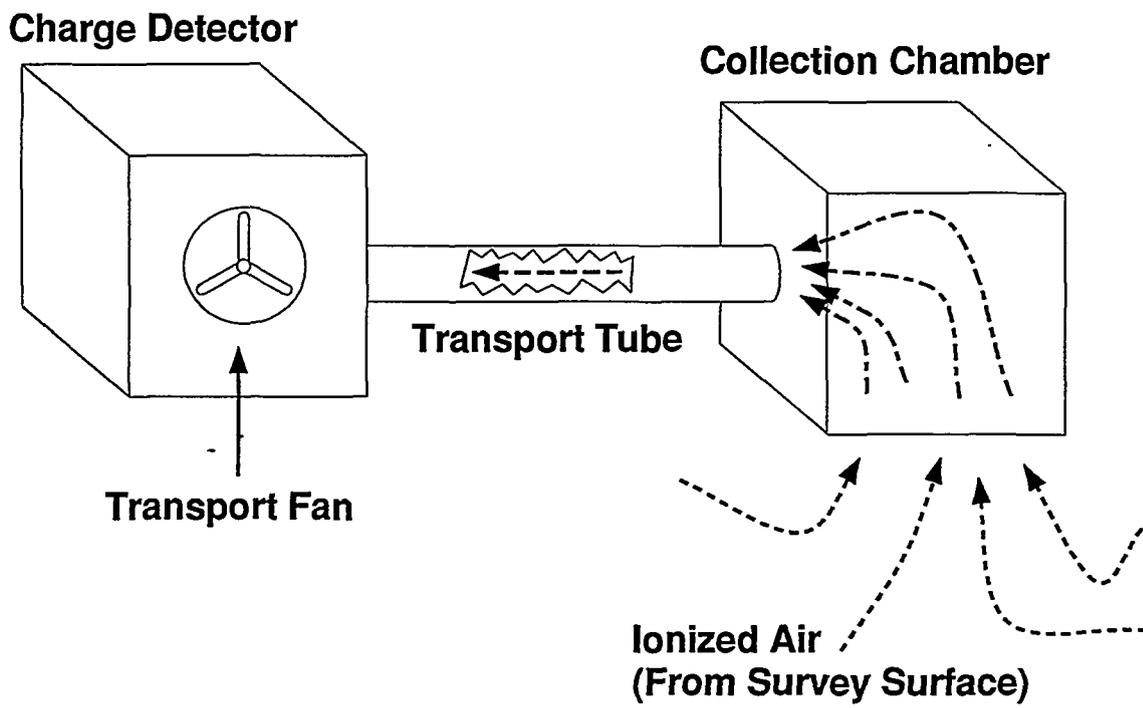


Figure 2-1. Illustration of the basic LRAD principle of operation

LRAD instruments have been demonstrated for a number of applications. These include:

1. Survey of items for uncontrolled release. The item of interest is placed inside a sensitive volume, and the ionized air is detected. If the item is placed on a mesh shelf arranged for full airflow around all sides, the item can be completely surveyed by drawing ionized air from all exposed contaminated surfaces in one measurement. LRAD instruments have been demonstrated with adequate sensitivity to meet requirements for unrestricted release from a radiological controlled area.
2. Surface monitoring in the field. A soil-surface monitor (SSM) powered by a rechargeable battery for field site-characterization studies of surface-soil contamination has been demonstrated at the Fernald, OH, site and other sites. This instrument has an active detection area of one sq. meter, and is configured for tractor mounting. This particular SSM does not use airflow; rather, ions are attracted to a collection grid operated with a high applied electric potential. It has a wide and linear operating range.
3. Monitoring pipes and ducts for internal surface alpha contamination. The instrument airflow is drawn from the pipe or duct of interest.
4. Monitoring absorbent material (such as filter paper) used to wipe a surface in a surface smear survey to check for removable contamination. The wipe material is placed inside a monitoring chamber and the resulting ionization is measured.
5. Personnel hand and arm monitoring. The person being monitored for alpha contamination inserts a hand (or hand and arm) into a monitoring chamber.

Whole-body contamination monitoring has been considered by the developers, but implementation of this application has been slower than other applications.

LRAD technology is being commercialized by Eberline.

2.3.2 Remote Source Detection

Special Technologies Laboratory has demonstrated a ground-level, remote ion-collector device for detecting ions transported through the air from an ionizing radioactive source. (Quam, 1995) This was a special instrument, for a specialized application, involving transport of ionized air for distances significantly larger than required for facility D&D, but it illustrates the feasibility of transporting ionized air several meters, if necessary to reduce operator exposure.

2.4 Laser-Induced Fluorescence

Special Technologies Laboratory also has investigated the phenomenon of laser-induced fluorescence (LIF) as a potential means of remote detection of radiation-induced ionized air. (Franks, 1992) They reported detection of fluorescence in the 426-428 nm range from ionized



nitrogen, resulting from laser excitation at 391.4 nm, but concluded that this technique was not a likely candidate for detection of low level sources.

3.0 PROVISIONAL REQUIREMENTS

In order to obtain the full benefit of alpha discrimination in the presence of strong radiation background, the detection system must be capable of some form of remote or semi-remote operation in order to reduce operator exposure. Further, it is highly desirable that the technology be able to perform surveys quickly and thoroughly when D&D workers remove surface material (paint, debris, etc.) and thereby expose significant alpha-contamination. We have already expanded the emphasis of this study to include survey of rubble and rough surfaces to meet the important D&D application of quick and efficient surveys to determine when removal of concrete and other material has proceeded far enough that the remainder can be handled as normal commercial waste.

Table 3-1 presents the list of relevant generic D&D requirements for alpha counting in the presence of strong background radiation, in support of D&D operations. The list in Table 3-1 is not explicitly tailored to force the selection of LRAD-type technology. However, a technology that remotely detects alpha-ionized air is a natural fit to the key requirements of rejection of high background at the survey location and of survey personnel protection through remote or semi-remote operation.

The basic requirement given in Table 3-1 is the detection of alphas using a method that automatically discriminates between alphas and beta/gamma radiation.

Next, it is recognized that full operational benefit of such an instrument will not be realized unless it is capable of some form of remote or semi-remote operation.

The goal of expediting operational surveys has led to identification of the requirement for convenient performance of surveys of rough surfaces, rubble, without significant risk of mechanical damage that would require repair of the alpha probe. Note that a system that efficiently and directly monitors rough surfaces and rubble for alpha contamination would not directly measure removable contamination, so it could not completely eliminate the need for time-consuming surface wipe surveys.

Full operational benefit would not be achieved from this alpha instrumentation technique unless it can respond quickly to freshly exposed contamination while operations are underway. The ability to perform continuous real-time monitoring to detect freshly exposed contamination appears to be very attractive.

Two limiting cases have been provisionally identified to permit specification of numerical radiation detection requirements. We see no reason why one particular instrument must be capable of operating over the entire range.

Table 3-1

Generic Requirements for Alpha Discrimination in Support of D&D Operations

1. The systems shall detect alpha-activity with rejection of high beta-gamma background at the survey location.
2. The systems shall be capable of remote or semi-remote operation, to protect the operator.
3. The systems shall be capable of surveying rough surfaces, rubble, etc., without significant risk of mechanical damage that would require repair of the detector probe.
4. It is highly desirable that the systems be capable of quickly detecting when operations remove surface material hiding significant surface alpha contamination.
5. The systems shall be capable of accurate detection of a wide range of alpha surface contamination source strengths. It is acceptable to achieve this range of operation by using different probes for different source ranges. Provisionally, the sources of interest range from:
 - 5.1 about 10 dpm/100 cm² (capable of performing surveys to meet the most restrictive measurement criteria to qualify items for unrestricted release) to
 - 5.2 a level of roughly 50,000 dpm/100 cm². (Capable of producing a dose of roughly 1 R/hr in a thin layer of ionized air above the emitter surface. See APPENDIX A.2)
6. It appears that the surface areas for a single measurement range from:
 - 6.1 about 50 cm² (consistent with the measurement criteria to qualify items for unrestricted release) to
 - 6.2 about one square meter (for survey of rubble and rough surfaces, to determine when demolition material no longer needs to be handled as alpha-contaminated waste).
7. The system designs and operating procedures shall take into account the expectation that the probe(s), and possibly the interior of the detector, can become contaminated during surveys.
8. The systems should have a capability for on-line measurement and subtraction of background at the location of the detector.
9. It would be helpful to be able to distinguish TRU alphas from radon daughter alphas at the survey location, but this already can be accomplished by semi-remote means using conventional health physics technology (e.g., air monitors, LC) with remote or semi-remote sampling.

At the low end, we wish sufficient sensitivity to be able to qualify tools and other objects for unrestricted release. Real tools and other objects presented for survey to qualify for release rarely have completely smooth, plane surfaces. An instrument capable of surveying alpha contamination without smears or careful scanning by an alpha probe would significantly expedite these surveys. Again, a system that efficiently and directly monitors rough surfaces and rubble for alpha contamination would not directly measure the quantity of contamination that would be removable, so it could not completely eliminate the need for time-consuming surface wipe surveys.

The high end detection requirement presented here is admittedly somewhat subjective. It is possible that we may identify an additional requirement for a remote, (possibly reduced-sensitivity) real-time alpha detector that would operate continuously, drawing air from a relatively large area, as a quick-response backup personnel protection instrument.

The specification of instrument sampling area numerical requirements parallels that of instrument sensitivity. At one end, there is a requirement for surveying tools and other objects for unrestricted release. At the other end, there is a requirement for efficient surveying of working areas. The selection of numerical requirements to quantify both of these types of area requirements is somewhat arbitrary, and the numbers must be considered to be subject to revision on the basis of operational experience.

Two requirements considered to be important for practical operations are the ability to decontaminate or replace components and the ability to efficiently subtract background on-line.

If radon and radon daughters are encountered in sufficient quantity to be of concern for personnel safety, their presence would be automatically detected by an alpha detector meeting the previous requirements. It could be helpful to the personnel planning operations to know if radon + daughters contribute a significant fraction of the alpha activity. We believe that existing instrument capabilities are sufficient to provide this discrimination.

4.0 EVALUATION PROGRAM

In view of recent demonstrations of a technology (LRAD) that appears to be very well-suited to D&D applications requiring alpha counting in the presence of high beta-gamma radiation background, and survey of rubble, the primary emphasis in testing and evaluation is directed toward practical implementation for D&D operations. Wherever possible, design feedback should be developed and documented.

For example, it is expected that the size and shape of the LRAD probe would affect the ratio of signal from air ion pairs generated by the short-range alphas to the signal from air ion pairs generated by the much more highly penetrating betas and gammas. This, in turn, would affect the sensitivity of the detector instrument to background beta-gamma activity.

A simple sketch illustrating this point is presented in Figure 4-1. The real situation would, of course, be more complex than the illustration. For clarity, the surface being surveyed is assumed to be plane. For simplicity, we assume that the surface emits alphas with a uniform source strength/unit area, and that the gamma-ray background dose rate in the air is essentially constant over a volume extending outward from the survey surface to many cm from this surface.

However, because alphas are highly ionizing, their path length in air is limited. As shown in APPENDIX A.1, ionization from the alphas is concentrated in a surface layer of air, about one cm thick. Hence, in the simplified example given in Figure 4-1, we show the highly ionized layer produced by the alphas to be one cm thick, containing I_α ion pairs/cm³ due to the alphas. The volume of air that is ionized by the gamma-ray background extends up many cm from the contaminated surface. Inside the probe, the gamma ionization is taken to be I_γ ion pairs/cm³, throughout the entire probe volume. For clarity, the probe is assumed to have vertical sides and a horizontal top, and it collects from a surface area, A. The top drawing shows a horizontal section of a probe that has its top located 10 cm above the survey surface. The thick lines denote the probe walls, and the surface being surveyed is indicated. Inside the one cm surface layer, the ionization per unit volume is equal to $I_\alpha + 10 I_\gamma$. Above this surface, the ionization per unit volume is only I_γ . The total number of ion pairs in this 10-cm high probe is equal to

$$A \times (I_\alpha + 10 I_\gamma).$$

On the other hand, if the top of the probe is located only one cm from the survey surface, as shown in the bottom drawing in Figure 4-2, the relative signal from the alphas is amplified. Again, the probe walls are shown by thick lines, and the surface being surveyed is indicated. In this case, the total number of ion pairs in the probe is equal to

$$A \times (I_\alpha + I_\gamma).$$

For the simple example in which $I_\alpha = I_\gamma$, the alpha signal from the tall probe is only about 5% of the total instrument signal. However, the alpha signal from the short probe is 50% of the total signal.

Table 4-1 presents the performance characteristics to be considered. It must be emphasized that an important aspect of the performance evaluation is testing the items given in Table 3-1 under practical conditions in order to determine what modifications and additions, if any, should be made in the requirements.



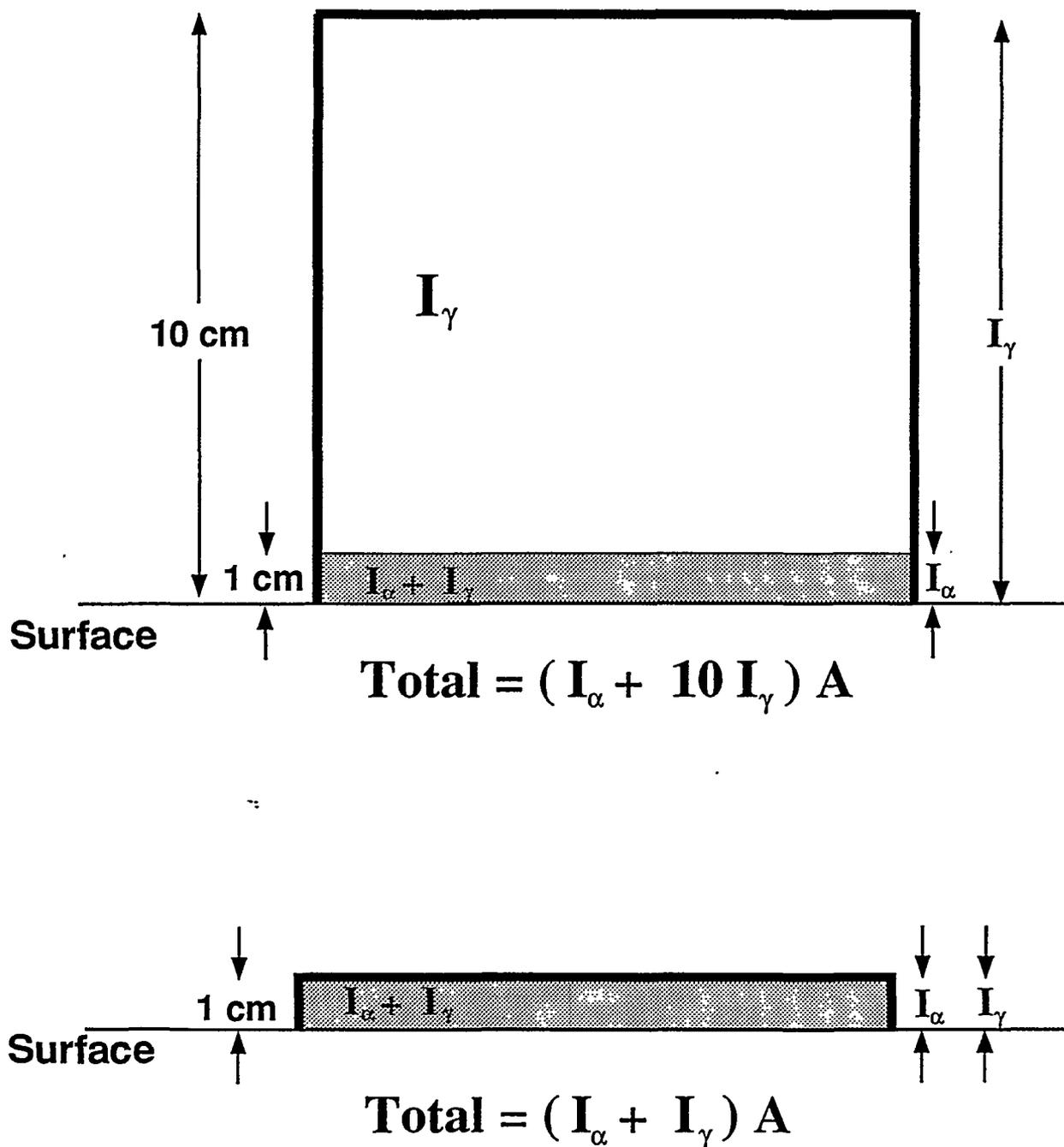


Figure 4-1. Illustration of effect of LRAD probe size and shape on the contribution of background gamma-radiation to the ionized air signal.



Table 4-1

Alpha Discrimination Performance Items to be Characterized

1. Discrimination against beta-gamma background. The ionization signal and the beta-gamma dose rate at the survey location, for a given alpha source and a range of beta/gamma dose rates.
2. Ease of handling. Record operator observations on the ease of handling for remote or semi-remote operations.
3. Practical operation at:
 - 3.1 low alpha contamination levels and at
 - 3.2 significantly higher alpha source strengths.
4. Practical operation for a range of sampling area setting(s) of interest for a single measurement.
5. Practical operation on rough surfaces. Record operator observations for operations on surfaces typical of facility operating equipment and of actual facility work station surfaces, and also concrete rubble. Describe and photograph test surfaces.
6. On-line measurement of background at the detector location. Measure background at this location and note feasibility of on-line subtraction of this background.
7. Practical aspects of probe operation where the probe and possibly the detector can become contaminated. Record operator observations on potential for detection of contamination, decontamination of probe and detector, and possible replacement of contaminated parts.
8. Noble gas contamination buildup. Practical aspects of buildup of background inside the instrument system due to radioactive noble gas retention by organics will be checked. Record operator observations.

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.

In addition, it should be noted that D&D applications include situations in which significant quantities of radon + daughters will be present. Therefore, attention will have to be given, at least for measuring the lowest ranges of alpha-radioactivity, to the retention of radioactive noble gas by organic materials. Although not widely reported, this phenomenon has been experienced in health physics operations involving radioactive noble gases. A good statement of the situation was given by Wahlig, et al. (1989):

"Although noble gases usually are not thought of as reactive or interactive with materials, experience with noble gas retention in a number of detector chamber materials have shown a surprising retention affinity. Polyethylene Marinelli beakers are observed to show a strong "memory" for noble gases because of the absorption of gases within the polyethylene structure. Background count-rate buildup in gross counting chambers has been observed to be caused by absorption of noble gases into gaskets, O-rings, or other organic seal materials within the chamber."

5.0 DISCUSSION

Discrimination of alpha activity in the presence of a high radiation background has been identified as an area of concern to be studied for D&D applications. Upon evaluating the range of alpha detection needs for D&D operations, we have expanded this study to include alpha counting of rough surfaces including rubble. We have developed provisional D&D requirements for instrumentation of this type (Table 3-1); and we also have generated the scope of a program of instrument evaluation and testing (Table 4-1), with emphasis on practical implementation.

In order to obtain the full operational benefit of alpha discrimination in the presence of strong radiation background, the detection system must be capable of some form of remote or semi-remote operation in order to reduce operator exposure. We have identified a highly promising technique (LRAD) for alpha discrimination in the presence of high radiation background. Neither the provisional requirements nor the evaluation and testing scope were expressly tailored to force the selection of a LRAD technology, and they could be used as a basis for studies of other technologies. However, a technology that remotely detects alpha-ionized air (e. g., LRAD) is a natural fit to the key requirements for rejection of high background at the survey location and operator protection. Also, LRAD appears to be valuable for D&D applications as a means of greatly expediting surface alpha-activity surveys that otherwise would need to be performed by time-consuming scanning with alpha detector probes and even more labor-intensive surface wipe surveys.

In developing a list of provisional D&D requirements and a provisional list of items to be evaluated, we have emphasized the practical aspects of implementing state-of-the-art alpha detection instrumentation technology. This includes specifying that the developers be given feedback from the evaluation activities. A specific example given of useful feedback is the size and shape of detector probes. It is likely that a range of instrument accessories will be required



in the field so that the operators can accommodate efficiently the range of conditions to be encountered during an actual D&D operation.

In addition, a true remote-viewing technique, laser-induced fluorescence (LIF), has been identified. Because of the limited amount of information available, the suitability of LIF for practical application to D&D operations is not clear at this time.

6.0 ACKNOWLEDGEMENTS

It is a pleasure to acknowledge the support extended to the Argonne Decontamination & Decommissioning Technology Center by the U.S. Department of Energy Office of Nuclear Energy Technology, and the guidance provided by the DOE program manager, S. Franks. Division NN-21 of the U.S. Department of Energy Office of Nonproliferation and National Security supported development of the new alpha-detection technologies reviewed in this report, under the direction of M. F. O'Connell. We also wish to recognize the coordination, encouragement, and technical comments supplied by T. J. Yule and D. R. Pedersen. D. B. Black, E. A. Rhodes, C. M. Sholeen, and G. S. Stanford also contributed valuable comments during the preparation of this report. Thanks also are due to Purdue University coop student R. Miller for his technical contributions, and A. Townsend for her essential secretarial services.

7.0 REFERENCES

ANL Environment, Safety & Health Manual, Chapter 5-9, Surface Contamination Surveys, p. 2, April 15, 1991.

L. Franks, STL, private communication, 1992.

J. D. Johnson, K. S. Allander, J. A. Bounds, S. E. Garner, J. P. Johnson, D. W. MacArthur, L. L. Sprouse, S. G. Walters, "Long-range alpha detector sample monitoring" Nucl. Instr. and Meth. in Physics Research, A 353, 486, 1994.

G. F. Knoll, "Radiation Detection and Measurement", 2nd Ed., New York: John Wiley, 1989, pp. 131-132.

D. W. MacArthur, K. S. Allander, J. A. Bounds, M. M. Catlett, and J. L. McAtee, "Long-Range Alpha Detector (LRAD) for Contamination Monitoring", IEEE Trans. Nucl. Sci, 39, 952, 1992a.

D. W. MacArthur, K. S. Allander, J. A. Bounds, K. B. Butterfield, and J. L. McAtee, "Long-Range Alpha Detector", Health Physics, 63, 326, 1992b.

W. Quam, STL, private communication, 1995.

T. M. Semkow, P. P. Parekh, C. D. Schwenker, R. Dansereau, J. S. Webber, "Efficiency of the Lucas scintillation cell", Nucl. Instr. and Meth. in Physics Research, A 353, 515, 1994.

B. G. Wahlig, D. M. Walker, M. R. Ghavi, and J. M. Palms, "Problems Associated with Routine In-Plant Radioactive Effluent Monitoring Systems at U.S. Light-Water Reactors", Nuclear Safety, 30, p. 29, 1989.



APPENDIX

A.1 Estimate of thickness of highly ionized surface layer of air above plane alpha-emitting surface.

We use a simple geometric model, as shown in Figure A-1. This model assumes that 5 MeV alphas are emitted isotropically from a plane surface covered by air at standard conditions of temperature and pressure (STP). The ionization track of each alpha is taken to be a straight line, 3.5 cm long, in the air. We consider only alphas emitted outward into the half-space above the emitter plane, and calculate the spatial location of alpha tracks for alphas emitted from a typical point on the emitter plane surface. We construct a half-sphere in the air with center at the point and radius of 3.5 cm. All of the alpha tracks end on the surface of this sphere. The requirement that alpha emission is isotropic means that the number of alpha tracks ending in a given region on the surface of the half-sphere is proportional to the area of that region, only. The direction of a track is defined by its angle with respect to the vertical line extending upward from the point. This angle ranges from 0° (vertical track) to, but not including 90° (horizontal tracks).

First, all alpha tracks emitted at an angle greater than

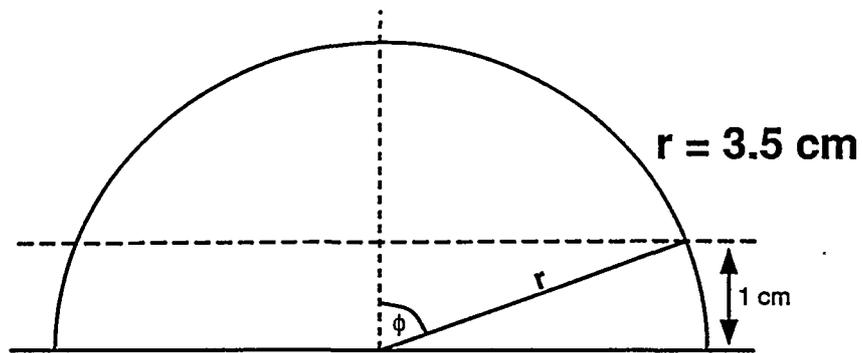
$$\arccos(1/3.5) = \arccos 0.2857 = 73.4^\circ$$

will be completely contained in the one cm thick layer of air immediately adjacent to the plane. This geometry is shown in the top portion of Figure A-1, which is a view through the half-sphere. Integrating the surface area of the half-sphere contained between the emitting plane surface and a parallel plane located one cm above the emitting plane shows that this area is given by

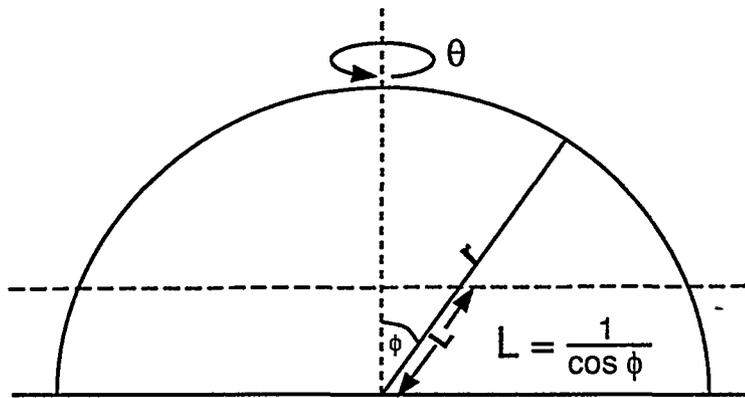
$$\text{Area} = \sin(16.6^\circ) \times \text{the surface area of the half sphere.}$$

That is, the area defined by alpha tracks that lie entirely in the one cm surface layer is 0.2857 x the surface area of the half-sphere. Thus, 28.57% of the alphas generate all of their ionization within the one cm air surface layer.

Next, we consider the case of alpha tracks emitted at angles less than 73.4° with respect to vertical. This case is illustrated in the lower portion of Figure A-1. Alphas emitted at an angle slightly greater than 73.4° with respect to the vertical will be almost completely contained within the one cm surface air layer. As the alpha track approaches the vertical direction, the portion of the track length inside the one cm surface layer, L , decreases to one cm, that is, a fraction of $1/3.5 = 0.2857$ of the full 3.5 cm track length. In the interest of clarity, we assume that the amount of ionization per unit length of alpha track is a constant. Numerically, L , the length of the track contained in the one cm air surface layer for a track emitted at a given angle with the vertical is a function of the emission angle, as shown in the Figure.



$$\phi = \cos^{-1}(1/3.5) \approx 73.4^\circ \approx 1.2810 \text{ radians}$$



$$0 \leq \phi \leq 1.2810$$

$$0 \leq \theta \leq 2\pi$$

$$\text{Area} = \int_0^{2\pi} \int_0^{1.2810} r^2 \sin \phi \partial \phi \partial \theta$$

$$\frac{\text{Area}}{2\pi r^2} \approx 0.7143$$

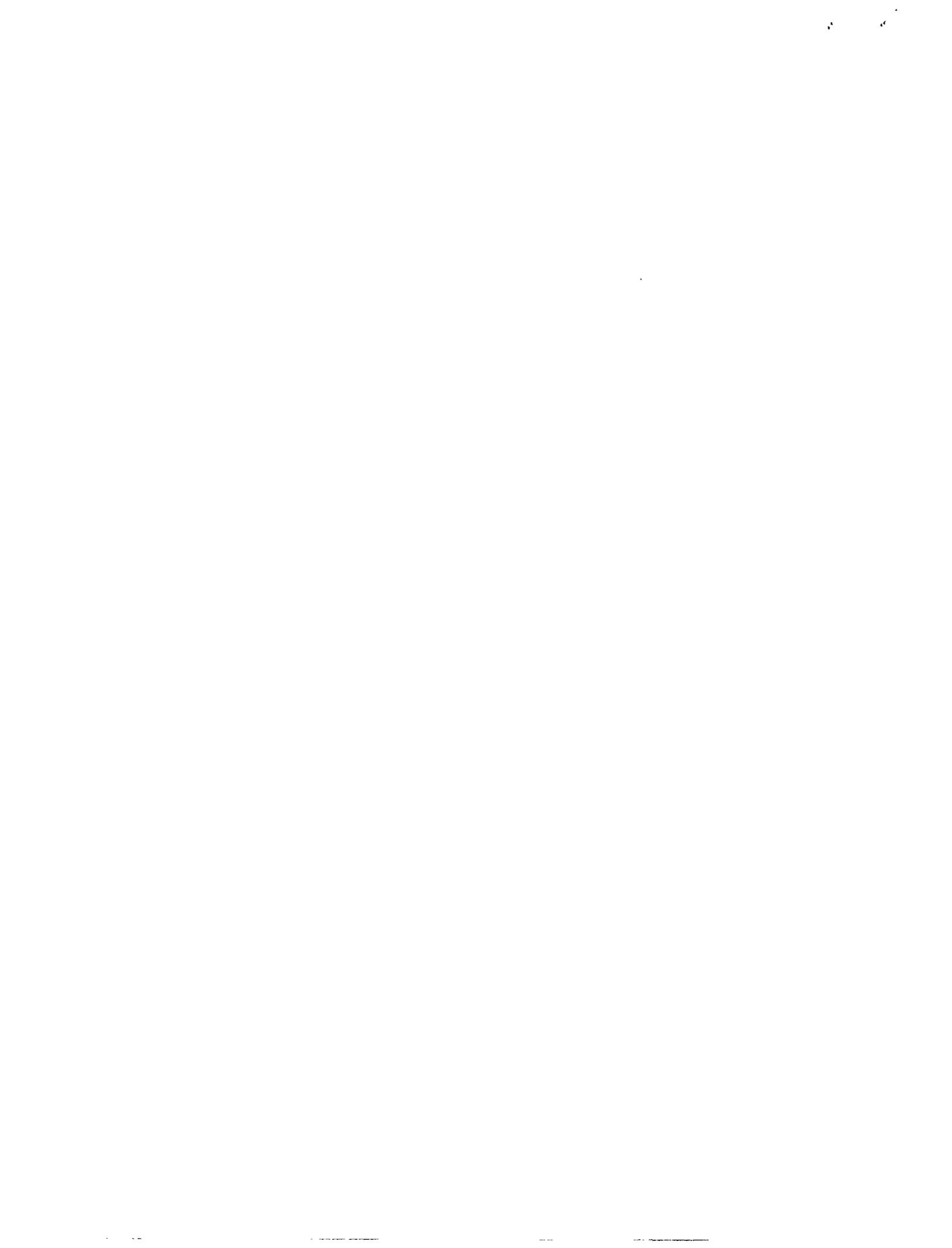
$$\text{Weighted Average} = \int_0^{2\pi} \int_0^{1.2810} r^2 \left(\frac{L}{r}\right) \sin \phi \partial \phi \partial \theta$$

$$\frac{\text{Weighted Average}}{2\pi r^2} \approx 0.3579$$

$$1 - 0.7143 = 0.2857$$

$$0.2857 + 0.3579 = \boxed{0.6436}$$

Figure A-1. Geometric model showing concentration of alpha ionization in a limited surface layer of air



Integrating, to obtain the average of total track length inside the one cm air layer for tracks that extend beyond the one cm air layer, weighted by the fraction of tracks emitted into this region of the half-sphere results in a

$$\text{Weighted average} = 0.3579,$$

for the tracks emitted at angles in the range between 0° and 73.4° with respect to vertical. This means that 35.79% of the total ionization generated by tracks from the point source is generated within the one cm surface layer of air by those tracks which extend above the surface layer.

Adding this ionization fraction to the fraction from those tracks lying totally within the one cm layer of air results in a total ionization fraction for all angles of emission of

$$\text{Total ionization fraction} = 0.2857 + 0.3579 = 0.6436.$$

That is, this model shows that 64.36% of the ionization produced by the point source is generated in the one cm surface layer of air.

In reality, the ionization per unit length of alpha track increases as the end of the track approaches; near the end of the track this linear ionization increases to about twice the value at one cm. Thus, the fraction of ion generation inside the one cm air layer for tracks that extend past that layer is not proportional to the relative track lengths inside the one cm layer, so that the fraction of ion generation inside that layer from the tracks that extend beyond the layer is less than the weighted average of 35.79 % calculated using this simplified model.

In view of the simplifying assumptions used in this analysis, we will adopt a generic scoping estimate that 50% of the alpha-radiation-induced-ionization occurs within a one cm air layer adjacent to the alpha-emitter plane.

A.2 Estimate of relationship between alpha dpm/cm² and dose rate in R/hr in the highly ionized air surface layer.

We will define 1 R/hr as the radiation dose rate that generates 2.08×10^9 ion pairs per hour per cm³ of air at standard conditions of temperature and pressure. For a dose rate of 1 R/hr, this yields $(2.08 \times 10^9)/(3.6 \times 10^3) = 5.7 \times 10^5$ ion pairs/second within a 1 cm cube of air. To estimate the dose-equivalence of alpha-rays in air, we use the approximate relationship from A.1 above that on the average, one 5-MeV alpha ray will expend 2.5 MeV in generating air ions within the "first" one cm layer of air above the contaminated surface. Using the datum of 35 eV/ion pair, this means that one average alpha will generate about 7.1×10^4 ion pairs within the one cm layer of air. From this, a dose rate of 1 R/hr in the one cm air layer having one face defined by an alpha-contaminated surface would require an emission rate of about 8 alphas/second/cm² in a direction outward from the contaminated surface. To be consistent with health physics terminology this will be converted to disintegrations per minute (dpm), and

referenced to a surface area of 100 cm^2 . Thus, one R/hr measured in a nominal one-cm air layer above a surface alpha source is estimated to correspond roughly to a measurement of 480 dpm/cm², or about

50,000 dpm/100 cm².



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