DP -188

Instrumentation

AEC Research and Development Report

A CONTINUOUS MONITOR FOR AIRBORNE PLUTONIUM

by

D. C. Collins
Engineering Assistance Section
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Printed in USA. Price $0.20
Available from the Office of Technical Services
U. S. Department of Commerce
Washington 25, D. C.

253405 ✓

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Printed for
The United States Atomic Energy Commission
Contract AT(07-2)-1

ABSTRACT

A continuous monitor has been developed that can detect one MPC (maximum permissible concentration) of plutonium in air within ten minutes. Application details are described to assist other groups in adapting the equipment to their needs.

External Distribution according to
TID-4500 (12th Ed.)

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A CONTINUOUS MONITOR FOR AIRBORNE PLUTONIUM

INTRODUCTION

In operations involving plutonium, very small particles may become airborne and escape into inhabited areas. Since plutonium is extremely toxic, early detection of such an escape is imperative.

At present, the only practical method of detecting very low concentrations of plutonium in air is to collect the particulate matter from a large volume of air and measure the alpha activity of the sample. If filter paper is used to collect the sample, the naturally occurring radon daughters are also collected. Since the plutonium activity cannot be distinguished from the radon daughter activity by ordinary counting equipment, the daughter activity must be allowed to decay for several hours before an accurate determination of the plutonium concentration can be made.

The annular kinetic impactor, developed at Chalk River, effectively solved the problem of radon daughter interference. Although this device permitted samples to be counted immediately after they were collected, a continuous monitor with a short response time and of sufficiently simple construction for practical use in the field was still desirable. This report describes the development of such a monitor.

Since it is known that groups at other sites are concerned with the same problem, the report also includes application details and background information which may assist such groups in adapting the equipment to their needs.

SUMMARY

A continuous monitor has been developed that can detect one MPC (maximum permissible concentration) of plutonium in air within ten minutes and a burst of high activity within two minutes. The monitor, which operates for a period of one week without attention, uses a slit-type impactor to reject radon daughters by selectively depositing the heavier plutonium particles on a moving tape which carries them under a scintillation detector. Air is sampled at the rate of 92 cfm, and the collection efficiency for plutonium is 55 per cent.

DISCUSSION

This monitor is the result of the combination and extension of the design features of two impactors described by others. The continuous sampling feature is described by Baurmash et al.[1] An impactor designed to collect plutonium preferentially was developed by Tait[2]. A brief resume of the prior art is given in the Appendix.

METHOD OF DETECTION

Particulate matter from the sample air stream is collected by means of a kinetic impactor, a device which accelerates the air stream and then causes it to make an abrupt right-angle turn. The velocity and thickness of the air stream are fixed at values which allow most of the radon daughters to follow the turn, while a significant fraction of the heavier plutonium particles fail to make the turn and strike the adhesive surface of a moving tape. The tape, moving at a constant speed, carries the collected solids from the impactor to a scintillation detector, whose output is indicated and recorded continuously.

PHYSICAL ARRANGEMENT

Top and side views of the complete monitor are shown in Figures 1 and 2. Figure 3 is a sketch illustrating the internal arrangement and method of operation. Figure 4 is a photograph of the interior of the monitor, showing the actual appearance of the portions illustrated in Figure 3.

Significant design and performance details are listed in the Appendix.

CONSTRUCTION OF IMPACTOR

The slit nozzle (Figure 5) was formed from two-inch-ID brass tubing by driving a wedge into one end while applying heat with a torch. The slit was finished to the desired dimensions (0.308 x 2.75 inches) with an end mill. The ends of the slit were rounded for convenience in machining.

The cross-sectional configuration at points between the circular portion of the tube and the slit is not important, provided the transition is smooth and gradual.

For a given slit width, the minimum particle diameter, D, for which the impaction efficiency is 100 per cent may be computed from the equation*,

$$D = (4.58 \times 10^{-2})\sqrt{\frac{d}{\rho v}}$$

where d is the thickness of the deflected streamline (or 1/2 the slit width), ρ is the density of the particle, and v is the velocity of the streamline. In this case, where $d = 0.385$ cm, $v = 8700$ cm/sec, and $\rho = 15$ gm/cm^3, the computed value of D is 7.89×10^{-5} cm, or approximately 0.8 micron. The density value used is less than the density of metallic plutonium, since in most cases the plutonium would occur as some compound.

COLLECTING TAPE

The monitor was designed to use "Scotch" brand masking tape, three inches wide. The collection efficiency of this tape for airborne plutonium was measured with a Chalk River type impactor, and the average efficiency for a ten-minute run was found to be 74 per cent. Although other adhesives (notably silicone grease) have higher efficiencies, tape coated with these materials is not available commercially.

In theory, all particles greater than the computed minimum diameter will strike the collecting surface. However, the collection efficiency for these particles will depend on the adhesiveness of the collecting surface. For an impactor having a stationary collection surface, the efficiency is decreased by "loading" of the adhesive. This efficiency decrease is exponential and is given by

$$E_t = E_o e^{-E_o fat}$$

where E_t is the efficiency after a time t, E_o is the initial efficiency, f is the particle flux, and a is the effective cross-sectional area of the particle.

* For air at standard temperature and pressure. See Appendix for derivation.

It can be seen from the equation that, for particles in the micron range, the exponent of e remains quite small for the first few minutes and the efficiency changes only a few per cent.

If the adhesive surface is moved continuously, so that no increment of area remains under the impactor slit for more than a few minutes, the loading is negligible and the collection efficiency can be considered constant.

TAPE TRANSPORT MECHANISM

The primary considerations in the design of the tape transport mechanism were (1) to provide a means of advance that was positive and uniform, and (2) to provide a simple method of "threading" the mechanism with tape.

In the final design, the tape is advanced by means of the take-up reel. Although this method results in increasing speed as the tape builds up on the reel, it is the most reliable means of providing the force necessary to unwind the tape from the original roll. The maximum increase in tape speed is held to 6 per cent of the starting rate by making the reel diameter large compared to the thickness of the deposited layer of tape.

A small synchronous motor and a gear train are used to drive the take-up reel at a constant speed of 1/2 revolution per hour. The reel is 8 inches in diameter and is increased to 8-1/2 inches by the addition of one 60-yard roll of tape, giving an initial tape speed of 0.209 inch per minute and a final speed of 0.222 inch per minute. The running time for one roll of tape is 167 hours, or almost seven days.

The path of tape travel was laid out so that only the nonadhesive side is in contact with the idlers. The tape makes a right-angle turn over a 1/4 inch idler located between the impactor and the detector in order to decrease the probability of contaminating the detector screen by impact of the air stream. The length of path between these units is short, to minimize the response lag.

To simplify threading, the transport mechanism is mounted on an aluminum plate that is hinged to the side of the enclosure. The entire assembly can thus be swung out of the enclosure, clear of the impactor and the detector.

Threading the machine with tape is a simple matter, but removing it from the take-up reel is more difficult. At present, the tape is removed by cutting through all the layers of tape down to the reel and then pulling it off in a single slab.

INSTRUMENTATION

The counting apparatus, except the scintillation detector, was assembled from commercially available components. The output of the detector is fed through a cathode-follower to a Mark 15, Model II Radiation Counter Laboratory count rate meter. This meter was modified to include a two-minute time constant. The indicated counting rate is recorded by an Esterline-Angus strip chart recorder.

The scintillation detector was made by spraying a zinc sulfide screen directly onto the face of a three-inch Du Mont 6363 photomultiplier tube. No special light shield is provided for the photomultiplier; instead, the entire tape housing is made light tight. To prevent damage to the phototube, its high voltage circuit is opened by an interlock switch whenever the housing door is opened.

CONTAMINATION

If the machine is operated for several weeks in a highly contaminated atmosphere, the background may rise slowly due to contamination of the alpha screen. The background can be lowered by cleaning the screen with a soft camel's hair brush, but a better remedy is to replace the photomultiplier tube with one that has a new screen. A contaminated screen can be wiped off and replaced by spraying, thus enabling the tube to be used again. The background can be measured by turning off the blower for about ten minutes to allow clean tape to be drawn under the counter.

OPERATING CHARACTERISTICS

Background due to collection of the radon daughters was determined by running the monitor for several days in an atmosphere known to be free of long-lived alpha emitters. In all other respects, the test atmosphere was similar to the atmosphere of the process room in which the monitor was to be used. The maximum counting rate observed was 20 c/m, and the minimum was about 8 c/m. If necessary, the radon background daily variation can be considered in interpreting the monitor indication.

When the monitor was operated in a contaminated atmosphere it was discovered that most of the contamination occurred as discrete particles of relatively high activity. Previously, when a filter paper sample indicated a concentration of a few times MPC, the general assumption was that the activity was uniformly dispersed throughout the sampled atmosphere. The monitor has shown, to the contrary, that the total collected activity may be due to only a few particles, some of which have activities high enough to constitute a sizeable fraction of the maximum permissible body burden. During one run, in an atmosphere containing an average plutonium activity of less than 1 MPC, a single particle was collected that had activity of 0.001 μc, or 12-1/2 per cent of the MPBB*.

RESPONSE CALCULATIONS

The method of converting indicated count rate to air activity is given below. The observed count rate is proportional to the specific activity of the air being sampled:

$$(1)\ \text{Count rate} = KC$$

where C is the specific activity of the air sample and K is a proportionality constant determined by the physical characteristics of the monitoring system.

* Maximum permissible body burden for the lungs as given in Radiological Health Handbook, U. S. Dept. of Health, Education and Welfare, October 1955.

The constant K includes the following factors:

Impactor efficiency	- e_I
Impactor slit width	- W_I
Sample flow rate	- F
Tape velocity	- V_t
Effective detector area	- A_d
Detector efficiency	- e_d

These factors are related as follows:

$$(2)\ K = \frac{FA_d e_I e_d}{V_t W_I}$$

The equation relating the count rate to specific activity is then:

$$(3)\ \text{Count rate} = \frac{FA_d e_I e_d}{V_t W_I} C$$

This relationship is correct as long as the maximum detector width is less than the impactor slit width. When the sensitive surface is wider than the slit, Equation 3 can be used if the area which is not directly over the deposited particle band on the tape is subtracted from the total area.

Sample Calculation for 1 MPC Air

For the monitor described in this report, the constants in Equation 3 have the following values:

e_I = 55 per cent (measured by comparison with filter paper samples)

F = 92 cfm (measured with a velometer at intake)

A_d = 4.91 sq. in. (2.5 in. sensitive diameter)

e_d = 33 per cent (measured with a standard source)

W_I = 2.75 in.

V_t = 0.216 in./min. (mean tape velocity)

C (1 MPC) = 2×10^{-12} μc/cc or
12.5×10^{-2} disintegations/min./cu. ft.

The count rate for 1 MPC air will be:

$$\text{Count rate} = \frac{(0.55)(92)(4.91)(0.33)(12.5 \times 10^{-2})}{(2.75)(0.216)}$$

$$= 17 \text{ counts/min.}$$

D. C. Collins
Engineering Assistance Section

APPENDIX

PRIOR WORK

The impactor used in the monitor described in this report is based on the cascade impactor, developed originally in England. In this device, several slit-type nozzles of decreasing size are connected in series to obtain particle size separation.

The Chalk River annular impactor uses the fact that collection efficiency depends upon particle size to discriminate against the radon daughters. The impactor dimensions are such that the collection efficiency is high for particles of diameter greater than about one micron, but drops off rapidly for smaller particles. Since the radon daughter activity is usually associated with particles of a diameter considerably less than one micron, the collection efficiency for these particles is comparatively low.

Several workers have developed continuous monitors employing a moving collecting medium. A device described by Hazard[3] used a moving strip of film and a jet impactor to collect atmospheric dust. The film then passed between a light source and a photoelectric cell to determine the amount of collected material. A device employing a moving strip of filter paper and a GM tube for the detection of airborne radioactivity was built by Kuper[4]. This machine had a very low sampling rate and was sensitive to radon daughters.

An atmospheric dust recorder that used a slit impactor was described by L. Baurmash et al.[1] The impaction surface was a rotating glass disk coated with silicone grease. The disk was removed and examined with a microscope after completing one revolution.

A stepwise continuous monitor has been under development at the Savannah River Laboratory. This instrument uses a Chalk River type impactor and a "Vaseline" coated film strip that is periodically advanced from the impactor to an alpha counter. It is designed to handle 120 cfm, has a high collection efficiency, and may be adaptable to requirements involving more precise measurement.

IMPACTOR THEORY

Consider a thin streamline flow striking a plane surface perpendicularly. Theoretically, the stream should divide in halves, each half making a 90° turn of radius, r. If v is the streamline velocity and m the mass of a particle in the streamline, then the magnitude of the centripetal force required to restrain the particle in the curve path is mv^2/r. The length of the 90° segment is $\pi r/2$, and the time t required for the particle to travel around the segment is given by $t = \pi r/2v$. If the particle is considered to be a sphere of radius R, its radial velocity V can be computed from Stoke's law. The result is

$$V = \frac{2R^2 \rho v^2}{9\eta r}$$

where ρ is the density of the particle and η is the coefficient of viscosity of the fluid in the streamline. The equation is valid for all streamlines if the thickness of the streamline is small compared to the mean radius of the turn.

The radial distance s traveled by the particle across the streamline during the turn is

$$s = Vt = \frac{\pi\rho}{9\eta} R^2 v$$

Note that s is independent of the radius of the turn.

The value of η for air at 18°C and 760 mm of mercury is 1.827×10^{-4} poise. Substituting this value gives

$$s = (1.91 \times 10^3)\rho R^2 v$$

If s is set equal to the thickness d of the deflected half of the streamline, and the substitution $D/2 = R$ made, the equation can be solved for the minimum diameter, D, of a particle that will strike the deflecting surface even though the particle was originally in the outer edge of the streamline. The result is

$$D = (4.58 \times 10^{-2})\sqrt{\frac{d}{\rho v}}$$

where D and d are in cm, v in cm/sec, and ρ in gm/cm^3.

The error in the result depends upon the correctness of the assumptions that (1) the particle is spherical, and (2) the particle movement across the streamline is so slow that turbulence does not set in (in which case Stoke's law for the retarding force does not hold).

IMPACTOR EFFICIENCY EQUATION

The equation relating the efficiency of an impactor after a time to the initial efficiency can be derived as follows: Consider a streamline flow containing uniformly distributed particles striking a surface that is coated with an adhesive. Assume the adhesive coating to be so thin that the particles are not absorbed in the adhesive, but instead just stick to the surface. As more particles stick, the available adhesive area decreases. The number of particles sticking in a small increment of time may be written as

$$\frac{dN}{dt} = E_o f(A - Na)$$

where E_o is the initial efficiency (or probability that a particle hitting the adhesive will stick), f is the particle flux (particles/second/unit area), A is the initial area of the adhesive surface, N is the total number of particles that have stuck, and a is effective cross-sectional area of the particle.

The total number of particles that have stuck after a period of time t is then

$$N = \frac{A}{a}(1 - e^{-E_o fat})$$

The efficiency at any time is the initial efficiency times the remaining adhesive area divided by the original area

$$E_t = E_o \frac{(A - Na)}{A}$$

or

$$E_t = E_o e^{-E_o fat}$$

DESIGN DATA

Impactor nozzle - See Figure 5
Air velocity exit impactor nozzle - 265 ft/sec
Spacing - Nozzle to tape - 3/16 in.
Spacing - Centerline of nozzle to near edge of scintillation detector - 7/8 in.*
Spacing - Scintillation detector to tape - 1/8 in.*
Tape speed - 0.209 in./min. initial
0.222 in./min. final
Scintillator detector - Zinc sulfide screen sprayed on face of 3 in. Du Mont 6363 photomultiplier tube
Effective diameter of scintillation detector - 2-1/2 in.
Collecting tape - "Scotch" masking tape - 3 in. width x 60 yd long

* Response time of monitor is less than that calculated from geometry using these dimensions because some alpha particles leave the tape surface at a forward angle so that the detector "sees" the dust sample before the tape is directly opposite the screen.

PERFORMANCE DETAILS

Net count rate from 1 MPC plutonium - 17 c/m*
Background due to radon Maximum - 20 c/m
Minimum - 8 c/m
Average - 15 c/m
Counting system background - 3 c/m
Total count rate - 35 c/m

* See "Response Calculation", page 7 of Discussion.

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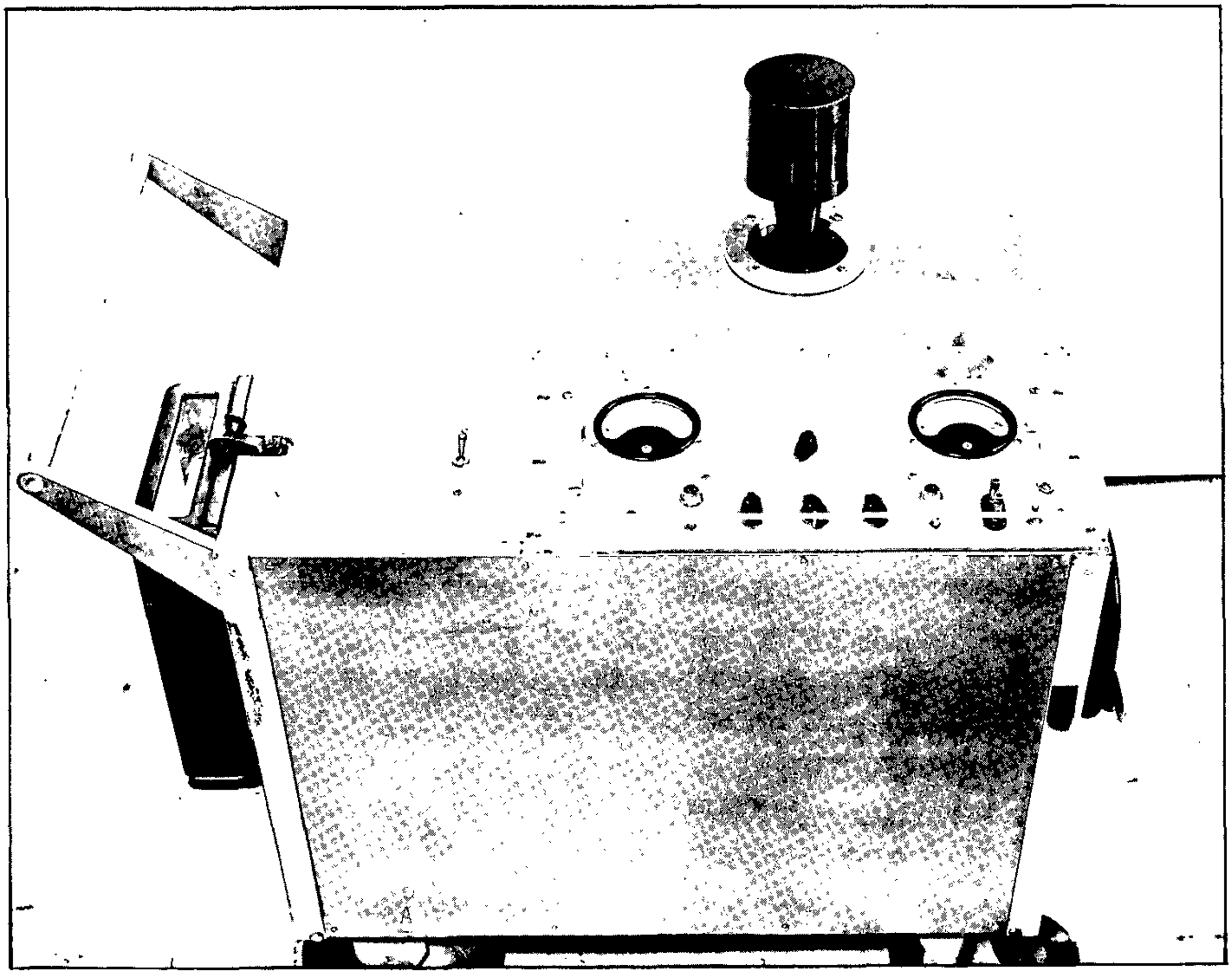
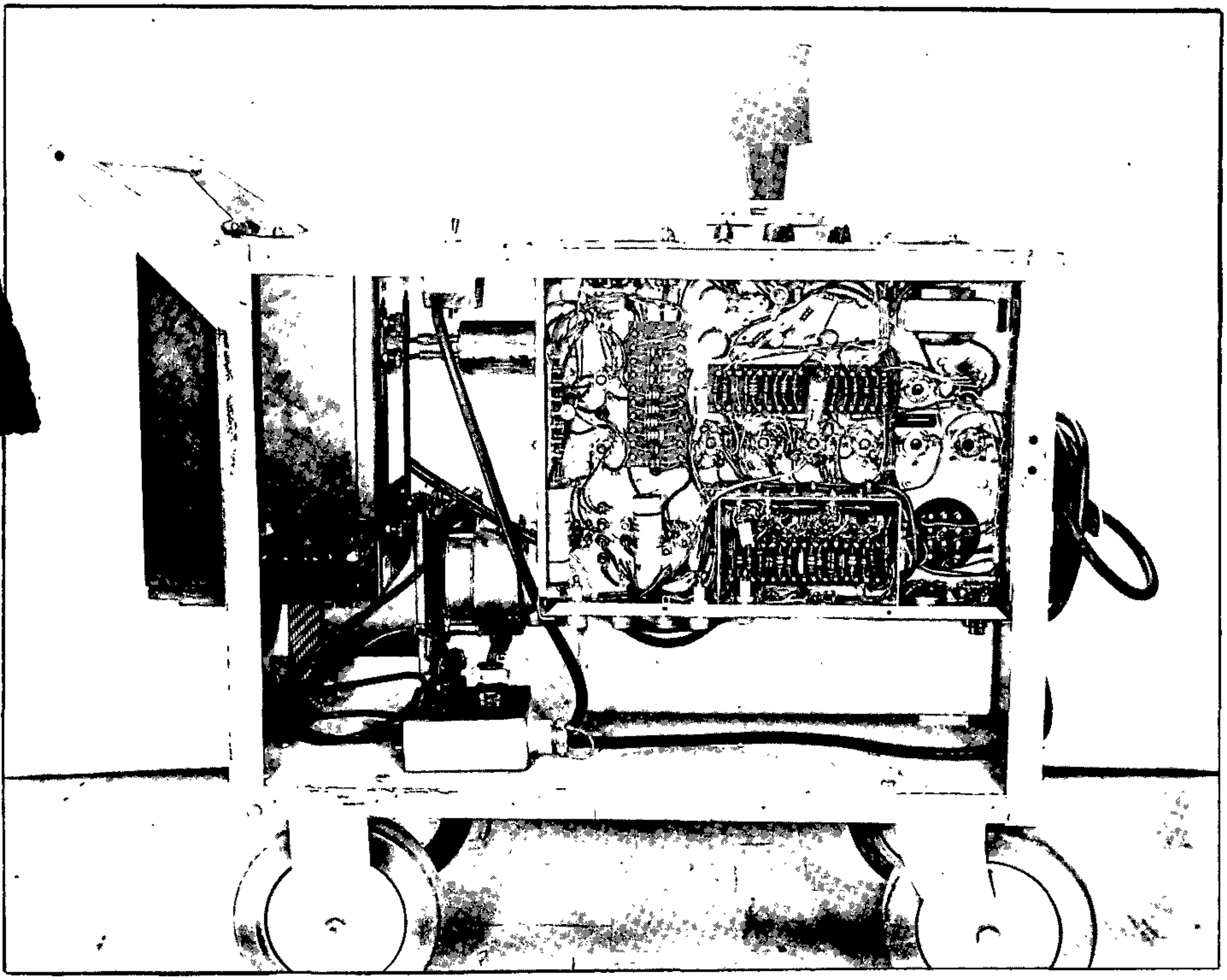
FIGURE 1



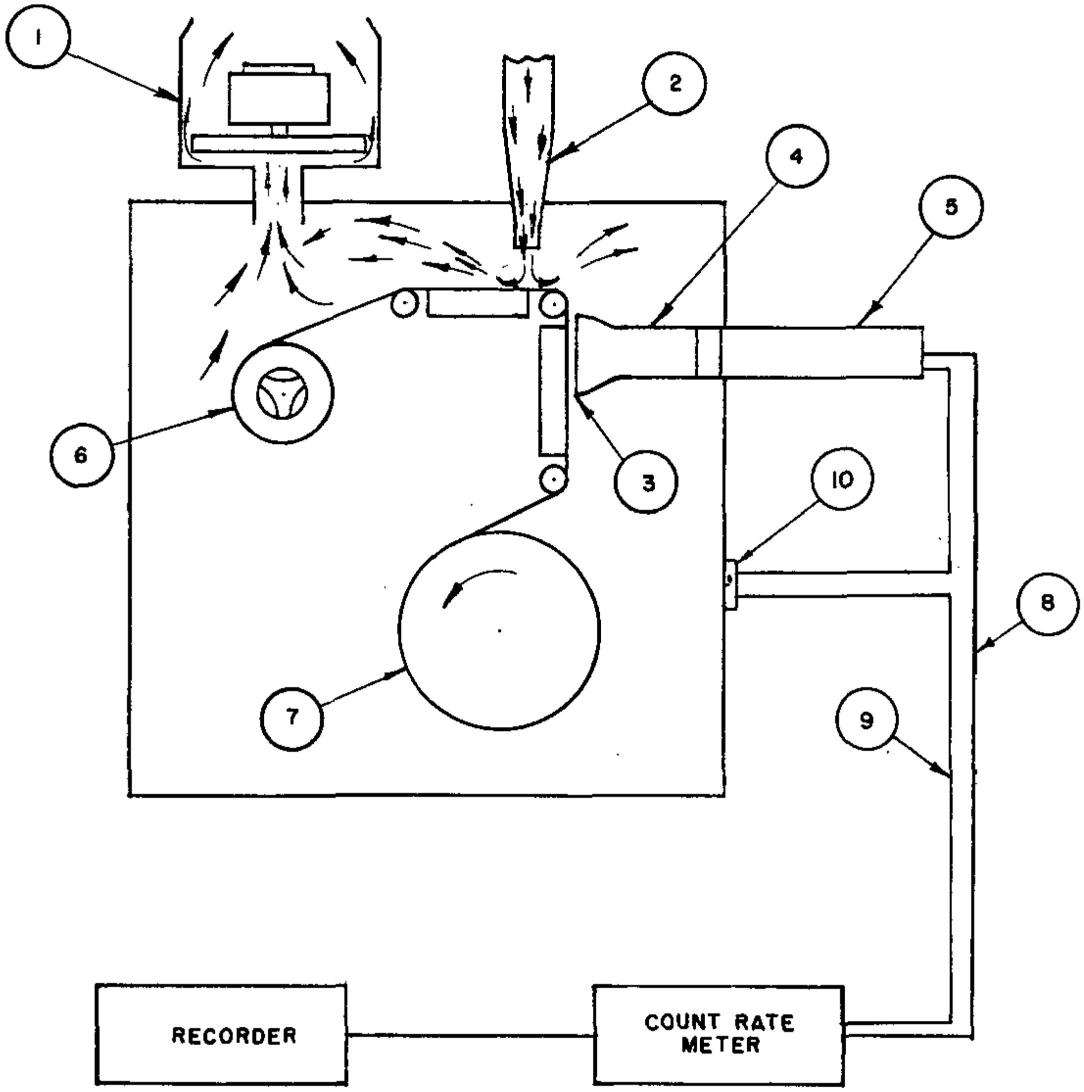
TOP VIEW OF PROTOTYPE MONITOR

FIGURE 2



SIDE VIEW OF PROTOTYPE MONITOR
(Side panels removed)

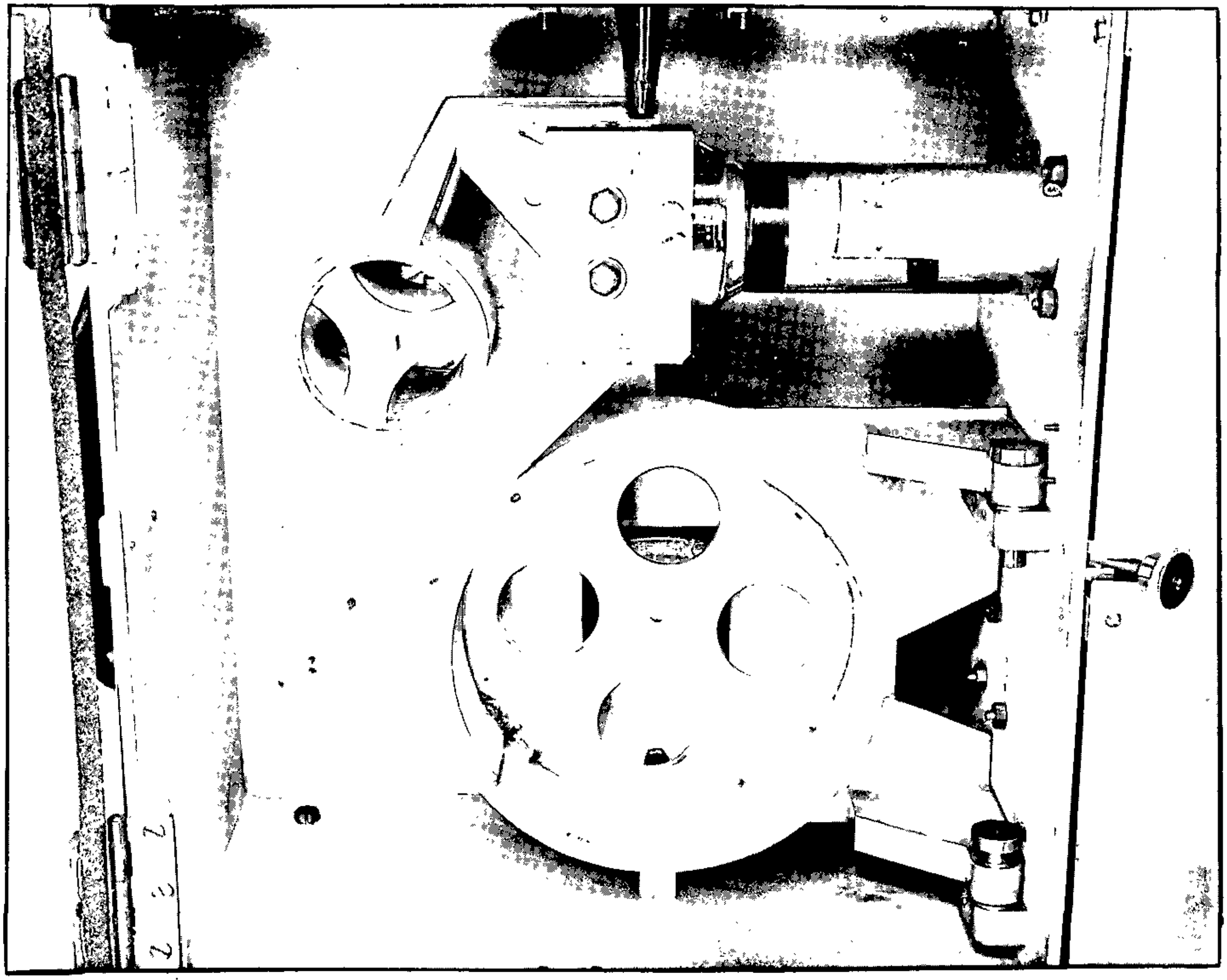
FIGURE 3



1. "STAPLEX" HIGH VOLUME AIR SAMPLER
2. IMPACTOR NOZZLE
3. SCINTILLATION SCREEN
4. P.M. TUBE
5. PREAMPLIFIER
6. MASKING-TAPE ROLL
7. TAKE-UP REEL
8. SIGNAL CABLE
9. HIGH VOLTAGE CABLE
10. HIGH VOLTAGE INTERLOCK SWITCH

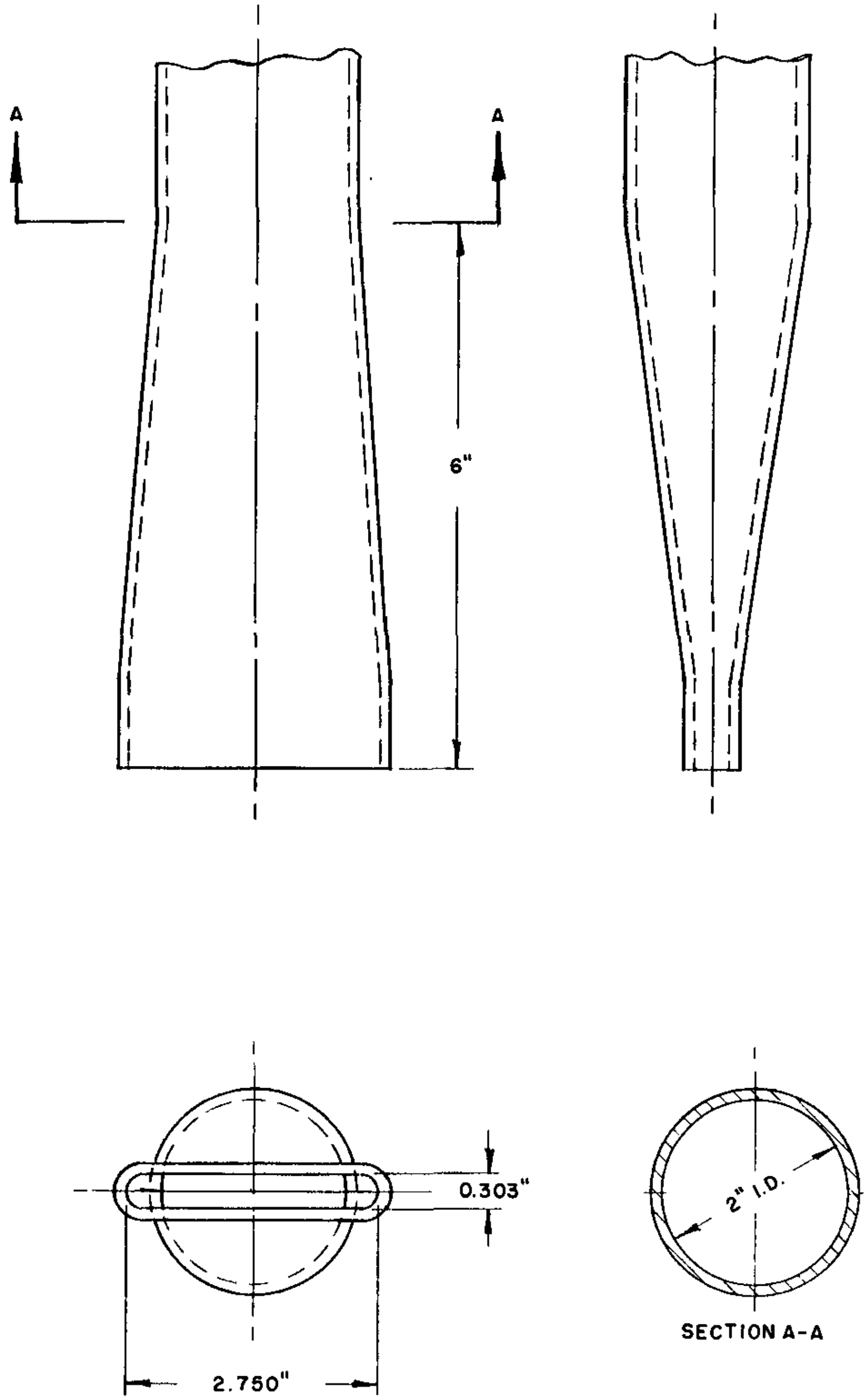
BLOCK DIAGRAM OF OPERATION

FIGURE 4



TAPE TRANSPORT MECHANISM, IMPACTOR, AND DETECTOR

FIGURE 5



IMPACTOR NOZZLE