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A MONITOR FOR LOW-LEVEL RADIOACTIVITY IN LIQUID STREAMS

by

E. C. Wingfield and P. R. Liller
Instrument Development Division

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E. I. du Pont de Nemours & Co.
Explosives Department - Atomic Energy Division
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ABSTRACT

A continual monitor for low-activity water streams is described. The monitor automatically collects constant-volume samples of the water, evaporates the samples to dryness, and presents the residues to two scintillation counters. It can reliably detect, within ten minutes, an alpha activity of 5.2×10^{-11} curie/ml, a beta activity of 7.3×10^{-9} curie/ml, and/or a gamma activity of 3.3×10^{-9} curie/ml.

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TABLE OF CONTENTS

LIST OF FIGURES AND TABLES

A MONITOR FOR LOW-LEVEL RADIOACTIVITY IN LIQUID STREAMS

INTRODUCTION

In a separations plant, it is necessary to monitor the amount of radioactivity present in certain cooling water effluents. These streams normally exhibit very little activity. In the event of a significant increase in activity the water is diverted to a retention basin.

A monitor was desired which would indicate continually and automatically the activity present in these coolant streams. It was specified that the instrument detect reliably alpha activity of 4×10^{-11} curie/ml and beta and/or gamma activity of 1×10^{-9} curie/ml. No such instrument was available commercially.

SUMMARY

A monitor was developed which measures out at least five constant-volume samples per minute of water, evaporates the samples to dryness, and presents the dry residues to two scintillation counters. One counter measures alpha activity; the other measures beta and/or gamma activity.

The monitor can detect an alpha activity (from natural uranium) of 5.2×10^{-11} curie/ml, a beta activity (from strontium 90) of 7.3×10^{-9} curie/ml, and/or a gamma activity (from cobalt 60) of 3.3×10^{-9} curie/ml.

DISCUSSION

DESCRIPTION

The monitor is an instrument that measures and records continually the amount of radioactivity present in the cooling water from a separations plant. Provision is made to sound an alarm should the activity exceed specified limits. Excessive activity requires diversion of the water into a retention basin.

The monitor is shown in Figures 1 and 2. The design of the mechanical apparatus was carried out by The American Machine and Foundry Co., while the specification and assembly of the electronics was done by the Savannah River Laboratory.

Water is continuously circulated to the monitor by a sump pump. Water samples are drawn from this line by a reciprocating metering pump, with an output per stroke that is adjustable from 1.0 to 1.5 cc. Aluminum tape is unrolled from a supply reel and "dimpled" by a reciprocating die. This feature led to the name "Dimple Water Monitor." A volume of water, constant within two per cent, is metered into these dimples, and then is passed under radiant heaters. The dry residue left after evaporation proceeds to two scintillation counters where the activity is measured. The dimples are then flattened between rollers and the tape is rewound on a take-up reel. The speed of the tape machine can be set at 312, 432, or 558 dimples per hour. The tape supply per reel is large enough to operate the machine continuously for at least 48 hours.

The sensing elements are a thallium-activated sodium iodide crystal for beta and gamma radiation and a silver-activated zinc sulfide screen for alpha particles. Each phosphor is "viewed" by a photomultiplier tube which supplies pulses to a corresponding scaler. Scaler, curve drawing attachment, and recorder are interconnected for automatic operation. A block diagram of the Dimple Water Monitor is shown in Figure 3.

Automatic operation is dependent upon a sequence of events. By means of the "preset count" feature, the scaler cuts off when a predetermined number of pulses have been counted. The curve drawing attachment then supplies a signal to the recorder. This signal is directly proportional to the logarithm of the activity. The recorder is a two-point instrument; one point indicates alpha activity and one point indicates beta and/or gamma activity. As the recorder prints, it resets the corresponding scaler, which in turn resets the curve drawing attachment. After a delay of five seconds, the curve drawing attachment starts the scaling operation, and the whole cycle is repeated.

The activity alarms are controlled by the recorder. There are two alarms for each recorded point, an "approach" and an "emergency." The emergency alarm is set to function when the maximum tolerable activity is obtained; the approach alarm is set at 50 per cent of this level.

PERFORMANCE

The Dimple Water Monitor was calibrated by means of liquid sources of known activity. The following data were obtained:

For the alpha particles of natural uranium one net count/min corresponds to 5.2×10^{-12} curie/ml.

For the beta particles of strontium 90, one net count/min corresponds to 7.3×10^{-12} curie/ml.

For the gamma radiation of cobalt 60, one net count/min corresponds to 3.3×10^{-12} curie/ml.

The background of the alpha probe is fairly constant at 0.5 count/min. The background of the beta-gamma probe exhibits considerable fluctuation. The average during any one day, taken over a large number of independent counting intervals, varies from 170 to 200 counts/min.

The minimum activity levels that can be detected reliably by the Dimple Water Monitor or a similar instrument depend upon a reasonable compromise among several factors:

1. Counter efficiency

2. Sample size

3. Background level and variations therein

4. Number of counts, which for good statistical reliability should be large

5. Counting time, which for maximum speed of response should be short

6. Frequency of equipment calibration which includes determination of background

7. Method of interpretation of the results

No provision is made in the Dimple Water Monitor for automatic recalibration and flexible compensation for varying background is not provided. The alarms are actuated on the basis of total counts (signal plus background). To minimize false alarms, allowances for variations in background are required in the selection of emergency alarm settings.

A practical compromise among all these factors was made as follows:

For beta-gamma counting

A net count to background ratio of five

A total preset count of 3000

For alpha counting

A counting time comparable to the beta-gamma counting time (at the alarm level)

A net count to background ratio of 20. The inferiority of the alpha counting statistics to beta-gamma counting statistics justifies a larger ratio.

With the calibration and background data of the monitor, the above requirements lead to emergency alarm settings equivalent to 5.2×10^{-11} curie/ml of alpha activity, 7.3×10^{-9} curie/ml of beta activity, and 3.3×10^{-9} curie/ml for gamma radiation; these figures are respectively 1.3, 7.3 and 3.3 times the activity levels originally specified. The corresponding counting times are 2.85 and 2.50 minutes, respectively, for preset counts of 30 and 3000. Because one sensing element is employed to detect both beta and gamma radiation, the alarm point is reached whenever the two types of radiation together yield more than 3000 counts in 2.50 minutes.

THE TIMING CYCLE

In order to retain control of the stream the water is fed into a delaying basin. The time required for passage through the delaying basin is about fifteen minutes, during which time the monitor must give an alarm if the activity exceeds the specified levels.

Since the instrument operates on the "preset count" principle, the maximum delay of an alarm occurs when the activity of the water rises abruptly from a very low level to a value slightly above the alarm level, shortly after the initiation of a counting cycle. Under these circumstances, the alarm will not be activated until the next cycle. At the fastest tape speed, the maximum delay is 9.8 minutes for alphas or 8.6 minutes for betas and gammas. At the slowest tape speed, the delay may be as long as 12.4 or 10.7 minutes. Higher activities reduce the counting times in the inverse proportion. The minimum dead time is approximately four minutes for alphas and three minutes for betas and gammas.

Table I presents an analysis of times required in the various steps. The figures represent the performance of existing equipment at activities that are slightly in excess of emergency alarm levels. All times given are maximum times required in the worst possible situation.

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BIBLIOGRAPHY

1. Tracerlab, Inc., CC-11 Curve Drawing Attachment Manual.

2. Tracerlab, Inc., CC-10 du Pont Scaler Manual.

3. Birks, S. B., Scintillation Counters. New York: McGraw-Hill Book Co., p. 51 (1953).

APPENDIX

DETAILED DESCRIPTION OF THE COMPONENTS

Scaler

A Tracerlab CC-10 scaler is used. The scaler can be "stopped" and "started" remotely.

Recorder

A 50-millivolt, two-point, synchroprint Brown recorder is used. Switches are provided to reset the scaler and to activate either the approach alarm or the emergency alarm.

Probes

The scintillation probes used in the monitor are General Electric stabilized counting room probes, Catalog #122C255G1. Each probe is fitted with a special sensing element that is suitable for its particular job. Each probe consists of an RCA 5819 photomultiplier tube, a variable gain amplifier, and a cathode follower that drives a coaxial cable which transmits signals to the scaler. The amplifier increases the output signal of the photomultiplier up to 0.25 volt, the value required to trigger the scaler. All these units are contained in a stainless steel cylinder 2-3/8 inches in diameter and about 13 inches long. Two cables connected to the scaler furnish the probe with all necessary voltages.

Detection of Alpha Particles

The sensing element for alpha particles is a screen which fluoresces under alpha-particle bombardment. The screens are plastic sheets coated with silver-activated zinc sulfide. They were prepared in the laboratory. A suspension of ZnS(Ag) in toluene and "Q-dope" was sprayed on plastic plates. The toluene dissolved a thin layer of the plastic and allowed the ZnS(Ag) particles to sink to the bottom of the layer. Then the toluene evaporated and the plastic hardened. The ZnS(Ag) particles were left firmly embedded just below the surface.

Screens prepared in this manner have been shown to be almost 100 per cent efficient. A counting efficiency of 51.4 per cent was attained for a counter using such a screen with a plutonium source in direct contact. The maximum counting efficiency, including back-scattering, was 52 per cent under these conditions. Birks(3) states that the optimum thickness of the deposited ZnS(Ag) layer is between 10 and 25 mg/cm^2. Laboratory experiments confirmed these figures.

To protect the photomultiplier from ambient light, the screen was covered with a single layer of aluminized "Mylar," 0.0003 inch thick. Thirty per cent of the incident alpha particles are stopped by this light shield.

The geometry of the installation requires the alpha particles to traverse approximately 3/8 inch of air between the source, that is the residue in the dimple and the "Mylar"-covered ZnS(Ag) screen.

The alpha head, as shown in Figure 1, consists of an elliptical enclosure whose sides slope inward to a circular shoulder into which the steel probe housing is fitted. The inside walls of the enclosure are painted with white paint of high reflectivity.

Detection of Beta and Gamma Particles

The beta-gamma probe is fitted with a thallium-activated sodium iodide crystal as the sensing head. NaI(Tl) is used since it is the best obtainable scintillator for gamma radiation and exhibits a relatively high efficiency in the detection of beta particles. The NaI(Tl) crystals, 1-3/4 inches in diameter and one inch thick, were supplied in canned form by the Harshaw Chemical Company. An aluminum light shield 0.010 inch in thickness covers the face of the crystal.

Alarm System for the Tape Mechanism

In addition to the activity alarms discussed previously, the tape-pulling machine has an alarm system that warns the operator when the machine malfunctions or when the tape needs replacing. The alarm is actuated under the following conditions:

Lack or interruption of power

Failure of cooling fan

Low supply of tape

Lack of tape motion

Breakage of tape

Failure of heater

These alarms actuate an annunciator system that is normally "On." False alarms may be caused by a failure of the alarm system. However, a malfunction of the equipment will always produce an alarm.

Activity Alarms

The preset count settings of the scalers were selected to be 30 for alpha detection, and 3000 for beta-gamma detection. The counting time is given by the relationship

$$\text{Counting Time} = \frac{\text{Preset Count}}{\text{Background + Net Counting Rate of Sample}}$$

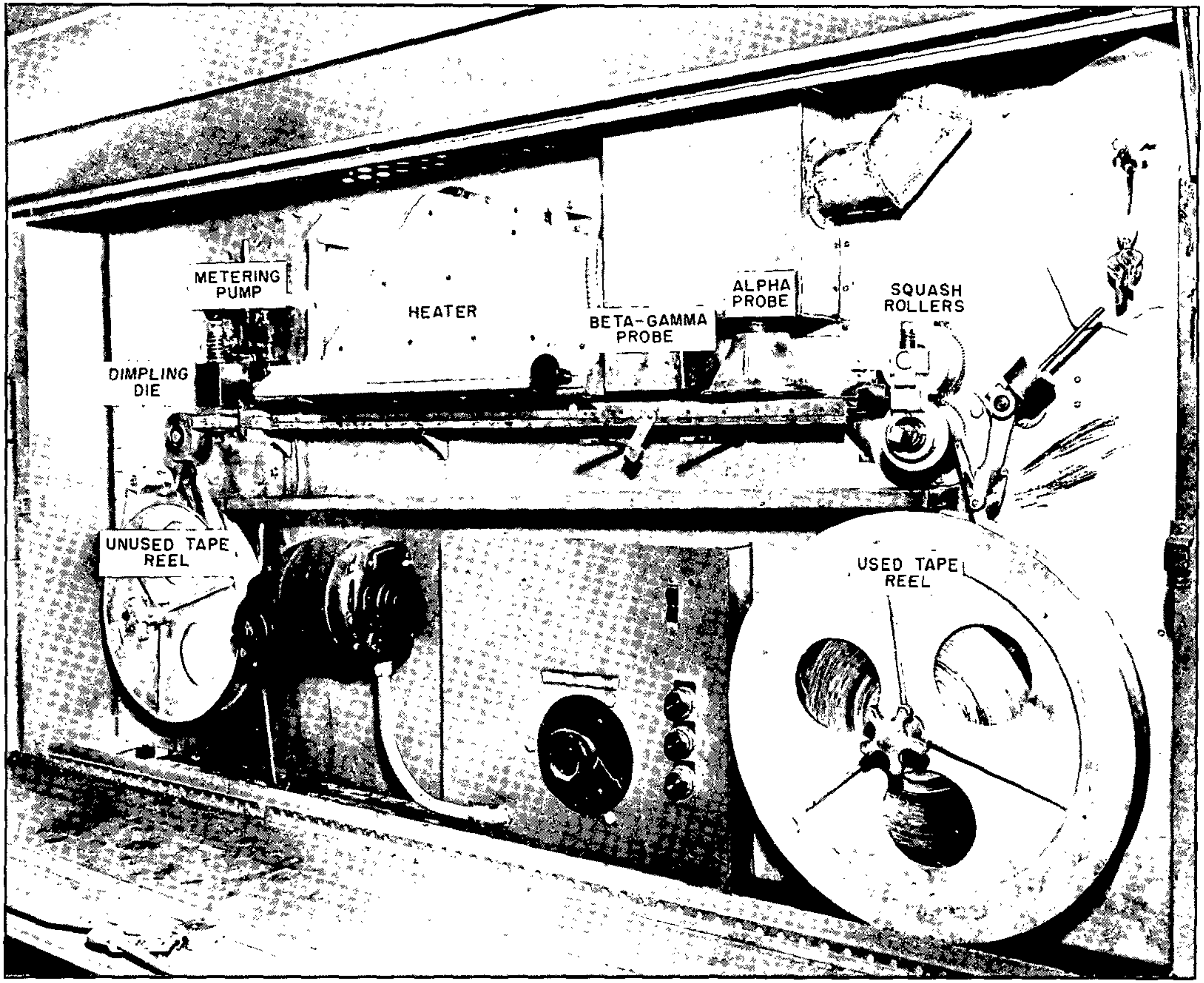
At the emergency alarm level, it amounts to

$$\frac{30}{0.5+10} = 2.85 \text{ minutes for alphas}$$

and $$\frac{3000}{200 + 1000} = 2.50 \text{ minutes for betas and gammas.}$$

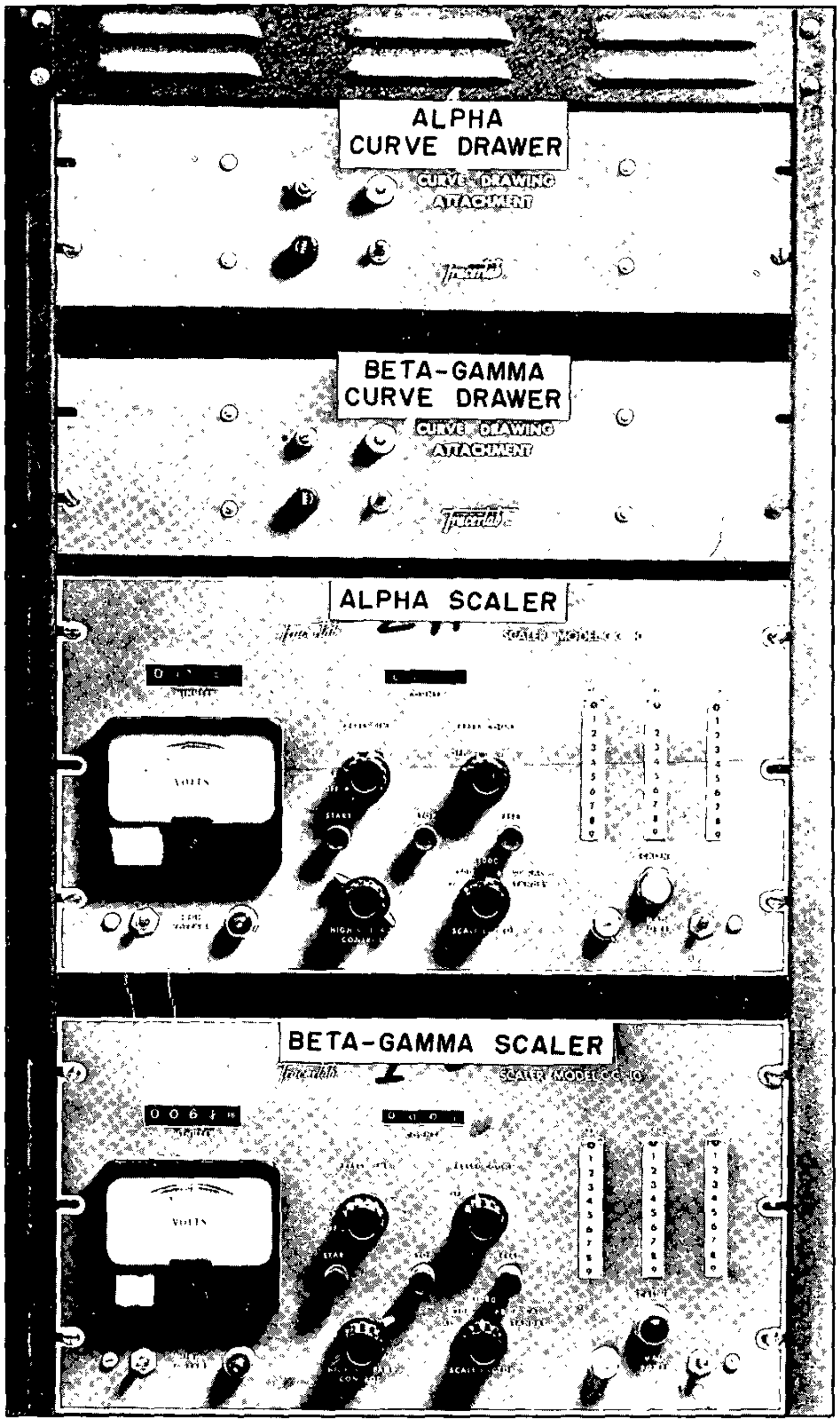
In the event of a sudden increase of alpha activity rising to slightly more than 5.2×10^{-11} curie/ml, it is possible for the monitor to require as much as 2.8 minutes to complete the preset count. If the 2.8 minutes plus the time elapsed in the present cycle, before the increased activity arrived at the alpha probe, is greater than 2.85 minutes, no alarm will be sounded. However, an alarm will be given during the next full cycle.

The same type of reasoning applies to the beta-gamma component for a sudden increase whose beta plus gamma activity is slightly greater than 1000 counts per minute. The maximum beta-gamma time that could be consumed in the previous cycle is 2.4 minutes, which is less than the alpha time of 2.8 minutes because of the higher beta-gamma background.



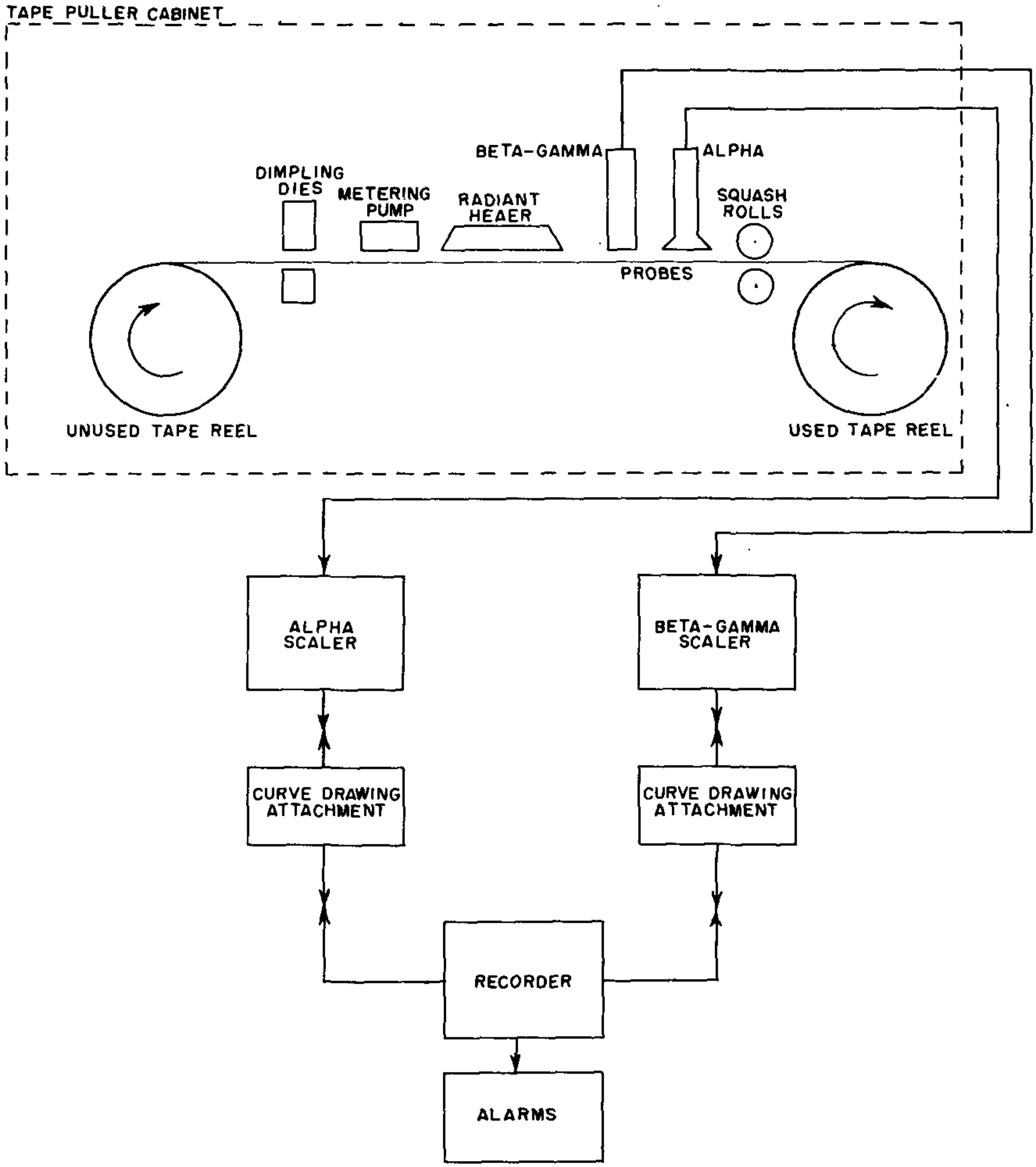
SAMPLING APPARATUS WITH CABINET DOORS OPEN

FIGURE 2



SCALERS AND CURVE DRAWERS

FIGURE 3



BLOCK DIAGRAM OF THE DIMPLE WATER MONITOR

TABLE I

TIMING CYCLE

For sudden increase to activity level slightly greater than value at alarm point:

	Alpha	Beta-Gamma
Emergency alarm point	10 net counts/min	1000 net counts/min
Preset count	30 total counts	3000 total counts
Travel time of water from sump pump to metering pump	10 sec	10 sec
Travel time of tape between filling point and probe for:		
Large gear	3.21 min	2.70 min
Medium gear	4.14 min	3.46 min
Small gear	5.80 min	4.85 min
Maximum scaling time remaining in previous cycle	2.8 min	2.4 min
Maximum time for recorder to print	20 sec	20 sec
Time for apparatus to reset	7 sec	7 sec
Counting time in first full cycle	2.85 min	2.50 min
Maximum time for recorder to sound alarm	20 sec	20 sec
Maximum total time to sound alarm:		
Large gear	9.82 min	8.56
Medium gear	10.75	9.32
Small gear	12.41	10.71