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A GAMMA-RAY COUNTING APPARATUS FOR EVALUATING THE FALLOUT REMOV--ETC(U)
MAY 77 G THOMSON, S SCHILLER, J GOSHORN

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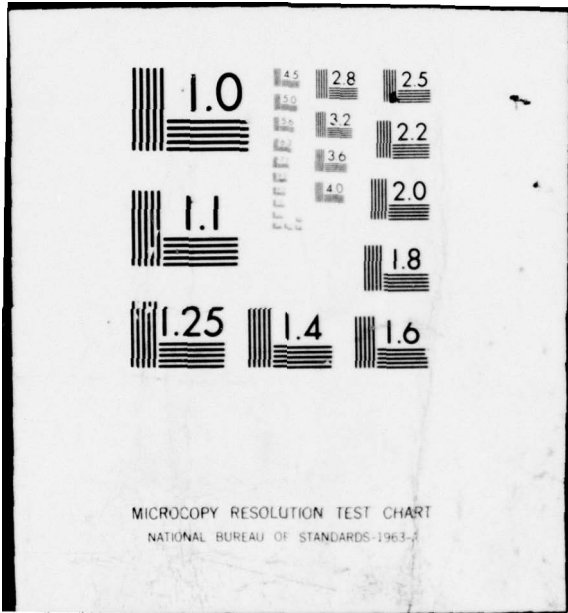
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MEMORANDUM REPORT NO. 2746

A GAMMA-RAY COUNTING APPARATUS FOR EVALUATING THE FALLOUT REMOVAL EFFECTIVENESS OF REVERSE-OSMOSIS WATER PURIFICATION SYSTEMS

G. Thomson
S. Schiller
J. Goshorn
E. Kaufman
C. Hollandsworth

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emphasizes counting efficiency, calibration accuracy, and a minimum of sample preparation. In tests on over four hundred samples using the system we measured the activity attenuation of streams of water following reverse-osmosis processing with error of a fraction of one percent. The accuracy of absolute dose level determinations was limited by the errors in the calibration standards at the highest levels and by counting statistics at the lowest levels.



* Was measured

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I. INTRODUCTION

The advent of nuclear weapons and power sources has presented us with the problem of providing potable water to human populations in radiologically contaminated environments, be they soldiers operating in a nuclear battlefield or civilians located near a nuclear disaster site. In response to this problem the Army and other agencies have developed several water purification systems. Among the most promising are those based upon the reverse-osmosis (R.O.) principle. In 1976 the Ballistic Research Laboratory joined with the Mobility Equipment Research and Development Center (MERDC) in a program to test the MERDC engineering prototype 600 gal/hr R.O. unit's ability to remove from water three important radioisotopes that occur in radioactive fallout: ^{131}I , ^{90}Sr , and ^{137}Cs . The present report describes but a small part of the test effort, namely the apparatus and techniques used to measure the amount of each of these isotopes in the test samples. Though many of the principles and techniques behind these measurements are not unique, they are being documented here in supplement form to provide a more complete picture of the measurements than is justified for inclusion in the final test report which, of course, is oriented toward R.O. system performance. We shall start with a short description of the test itself.

II. TEST PLAN

The R.O. test setup and its numbered components are shown in fig. 1. Each test was begun by adding a few millicuries of one of the subject radioisotopes in soluble form into a few thousand gallons of natural water (1). For safety and ease of detection the relatively short-lived gamma emitting isotopes ^{85}Sr and ^{134}Cs were substituted for ^{90}Sr and ^{137}Cs . The performance of the R.O. unit was, of course, unaffected by these substitutions. The dosed water was next filtered (2), and then passed through the R.O. unit itself (3). Tests were conducted on two parallel operating 600 gal/hr R.O. units, one with polyamide membranes (PA), and one with cellulose acetate membranes (CTA). R.O. processing separates contaminated water into two streams, one purified (4PA, 4CTA) and one more concentrated (5PA, 5CTA). Therein lies the principle advantage of R.O. - contamination does not accumulate within the system. Samples of raw dosed water, filtered water, purified water, and the concentrates were taken hourly at the points indicated by crosses. During the course of each five hour run all the products, both purified and contaminated concentrates, were mixed and were passed through an ion exchange unit (6). The ion exchange unit removed the isotopes from the water and fixed them into a bed of resins. Exchange processing continued until the water met the applicable environmental standards, whereupon it was discharged.

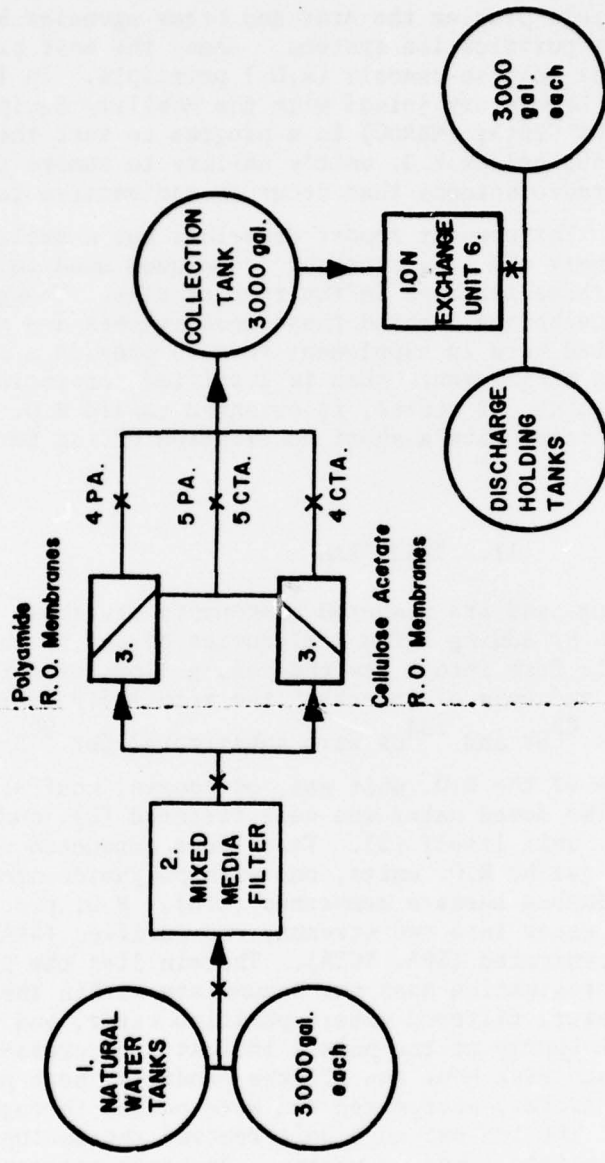


FIGURE 1. R. O. UNIT TEST SET-UP.

Test sample activities ranged from several hundred thousand picocuries/liter (pci/l) in the concentrate streams to a few tens of pci/l in the purified and discharge waters.¹ The system used to count our samples had to operate over this broad range of activities, had to be efficient enough to permit the determination of dozens of low level background and sample activities per day, and finally had to be accurate enough to measure the removal effectiveness of the R.O. unit to within a few tenths of one percent. For simplicity and ease of preparation the experimental design required that all samples would be of one liter size and would be counted intact, that is, they would not be boiled down or have their dissolved radioisotopes chemically separated prior to counting. The large number of samples that would be accumulated during this operation dictated that at least three separate, identical counting systems would be needed.

III. APPARATUS

The counting system consists of four parts, the detector, the counting electronics, the sample container, and the shield. Each will be discussed in turn.

A. Detector.

The three chosen radioisotopes ^{131}I , ^{85}Sr , and ^{134}Cs are all unstable. ^{131}I and ^{134}Cs decay by beta emission to ^{131}Xe and ^{134}Ba respectively, while ^{85}Sr changes to ^{85}Rb by electron capture. The daughter nuclei are all in excited states and all produce one or more gamma (γ) ray photons with characteristic energies in the range 0.1 MeV to 2.0 MeV.² Because γ -ray emission is common to all three isotopes and because γ -rays are sufficiently penetrating in water to permit efficient counting without prior evaporation, it was decided that our counting system should be based upon their detection. A one liter sample, at the lowest levels of sample detectability required of this experiment (about 30 picocuries/liter), produces less than one hundred γ -ray photons per minute. It is therefore necessary that the sensitive surface of our γ -ray detector subtend a large solid angle with respect

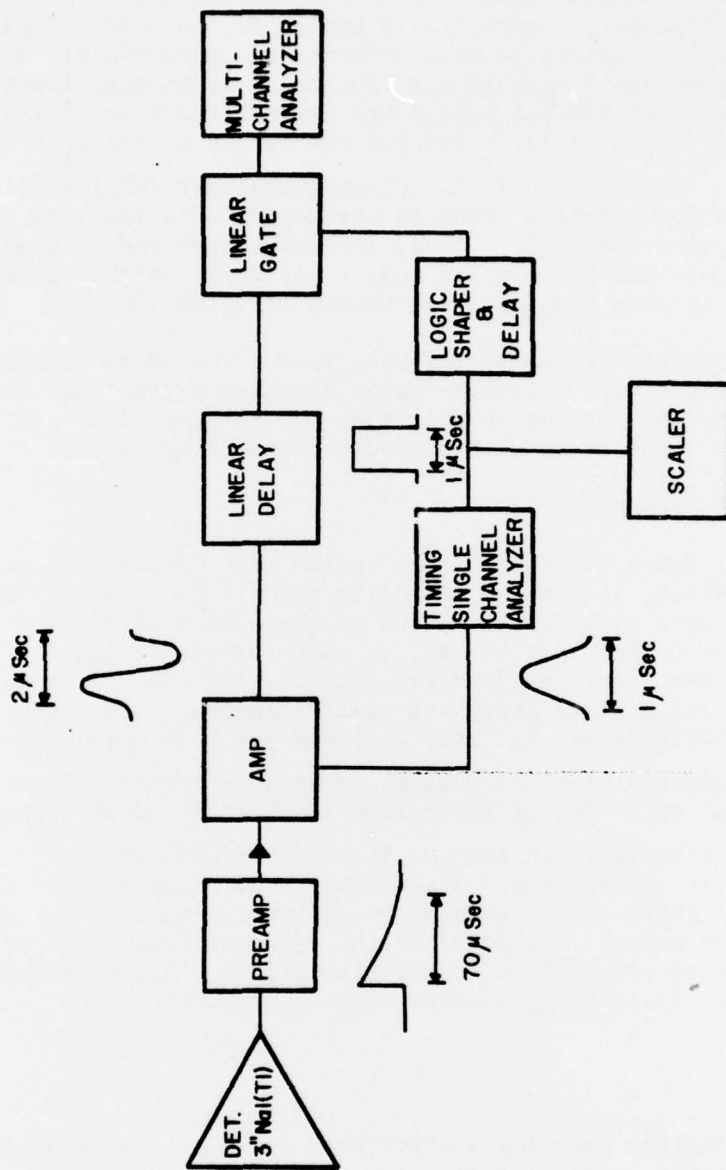
1. The maximum permissible levels for environmental discharge of the isotopes ^{131}I , ^{85}Sr , and ^{134}Cs are 300 pci/l, 100,000 pci/l and 9,000 pci/l respectively.
2. A complete description of all the decay products may be found in C.M. Lederer, J.M. Hollander, and I. Perlman, Table of Isotopes - John Wiley & Sons, New York (1967).

to the source. Any emitted gammas are likely to travel several centimeters in virtually any material before interacting with an atom to produce a detectable event. Efficient operation thus requires that the detector material be distributed with considerable depth along the γ -rays flight path. Both of these requirements are satisfied by scintillation detectors.³ A scintillation material produces a visible light output of intensity proportional to the energy of the interacting photon. Many such materials can be formed into large configurations. For the present purpose we employ cylindrical NaI(Tl) crystals that are 3" in diameter and are 3" long (Harshaw 12S12). The light output of the scintillator is gathered by a twelve stage photomultiplier and converted at the anode into an electron charge pulse whose magnitude is proportional to the light intensity. The phototube anode receives a bias potential of 1000 volts. Each of the other photomultiplier stages is supplied the appropriate potential for best gain and response characteristics via a resistive network. The photomultiplier and scintillator crystal are encased together inside a light and moisture tight aluminum jacket and are vertically mounted on an aluminum pedestal.

B. Electronics.

It is the function of the electronics to convert the charge pulse output at the photomultiplier anode into an electrical signal which permits recovery of the gamma energy information and, through this, the selection of only specific pulses for dose counting purposes. The system for attaining these goals is illustrated in fig. 2. The anode charge pulse enters a charge sensitive preamplifier (Canberra 805) that generates a corresponding tail voltage pulse as illustrated. The preamp output tail pulse has an amplitude that is proportional to the charge delivered to the anode and drives both a transmission line and the variable-gain main amplifier. The main amplifier (Ortec 410 or Tennelec 214) subsequently produces two RC circuit-shaped gaussian pulses - one unipolar positive going and one bipolar, positive-lobe leading. Each has an amplitude that is also proportional to its input height and bears a precise time relationship to the arrival of the fast leading-edge of the tail pulse. The unipolar pulse triggers a timing single channel analyzer (TSCA) (Ortec 420A or Tennelec 214) which in turn generates a square positive logic-pulse whenever the unipolar input amplitude falls between two limits e_1 and $e_1 + \Delta e_1$. The linear nature of the circuitry implies that a logic pulse will only occur when a gamma ray with energy in a proportionately large range E and $E + \Delta E$ is detected. The number of such γ -rays is easily determined by merely counting the logic pulses. A scalar for this purpose (Chronetics Nanocounter) is indicated. The problem remains of visualizing the particular feature or range of the

3. J.B. Birks, The Theory and Practice of Scintillation Counting, Pergamon, New York (1964).



ELECTRONICS

FIGURE 2. A block diagram of the detector and its associated electronics.

γ -spectrum that is selected by the settings e_1 and $e_1 + \Delta e_1$. To this end the bipolar pulse is taken, delayed (Ortec Model 489), and fed into a linear gate. The gate functions in two modes. When disabled, the gate allows all pulses, regardless of amplitude, to pass. When enabled, it only allows pulses to pass whenever an appropriately delayed logic pulse (Canberra 1455) appears simultaneously at another input. In the latter mode input bipolar pulses are transmitted only if the corresponding unipolar pulse falls between the limits e_1 and $e_1 + \Delta e_1$. Transmitted pulses are put into a multichannel analyzer (MCA) (Nuclear Data 160 series) which stores a count in its memory at a location proportional to the pulse amplitude. Thus, depending upon the gate mode, it is possible to accumulate and display counts vs. pulse height spectra for all pulse heights or only those falling within selected limits.

A separate counting device is included in our system to measure the elapsed counting time. All three gamma counting systems and the elapsed time counter are ganged so that they may be started or stopped together or in any combination with a single controlling switch.

C. The Sample Container.

The container which holds the water sample must possess at least three features. First, it must be optically thin to gamma rays; that is, it must not stop a significant amount of gamma rays within its walls. Second, for optimum efficiency, it must spread the liquid so that it surrounds the sensitive detector surface with even thickness. Finally, the material must be inert and easily cleaned. A design that is suitable for a cylindrical detector geometry has been developed by Marinelli.⁴ It consists of a cylindrical polyethylene beaker with a coaxial cylindrical well that is inset into its bottom and into which a detector may be inserted. In the one liter commercial version⁵ that we employed the wall thickness and diameters are adjusted so that the detector's top and sides are surrounded by an approximately 2 cm. thick layer of the sample water. The polyethylene construction allows the beakers to be cleaned and decontaminated with strong soap, acid and chelating solutions while being handled with only routine care.

D. Shield.

Low level radiation counting applications demand a reduction in the natural background counting rate. For this reason each counting cell is surrounded by a thick box-like lead container (interior

4. R.F. Hill, G.J. Hine, and L.D. Marinelli, Amer. J. of Roentgenol. Radium Ther, 63, 160-9 (1950).

5. Bel Art Products, Pequannock, NJ 07440, Model #F26850.

dimensions 24" high X 12" long X 6" wide). Each is mounted against a laboratory wall so that any externally generated gamma photon has to pass through at least 4" of lead or 2" of lead and a 10" high density concrete wall. A 4" thick lead lid was fashioned for each cell which can be easily placed or removed with the aid of specially constructed portable lift. The possibility of accidental liquid contamination of the shield is reduced by lining each cell with a plastic film. We found that the background rate of all detectable gammas drops to 3% to 5% of its unshielded values when the shield was in place, a reduction that is more than adequate for our purposes.

IV. CALIBRATION PROCEDURES

Two separate types of calibration operations must be accomplished prior to the taking of unknown sample dose data, namely the setting of the gamma energy acceptance window and the compilation of an accepted counts vs. dose curve. Because of variations in counting system gains the window must be set individually for each isotope in each of the three detector systems. The added variations in Marinelli beaker geometry mandate that the counts vs. dose calibration must be performed for each combination of isotope, detector and Marinelli beaker used.

Radioactive standards of each isotope in liquid form⁶ were obtained for the calibration runs. The dose errors in the standards were 2.9%, 4.5%, and 1.1% for ^{131}I , ^{85}Sr , and ^{134}Cs respectively at a 99.7% level of confidence. These individual solutions were accurately diluted with distilled water in such a way that a series of products were obtained, each with 20% of the activity per liter of its parent solution. The dilution process was continued until the activities of the various solutions spanned the range of experimental interest for each isotope. About two liters of each were prepared.

To set the TSCA energy acceptance window 1000 grams of the test solution at about 100,000 pci/l activity were added to a Marinelli beaker, which was then in turn placed over a detector and inside a shield. The detector bias was then applied and a multichannel analyzer spectrum of γ -ray counts vs. pulse height was obtained with the gating system disabled. Examples of such spectra are shown in figures 3, 4, and 5 respectively. Reference to those figures shows that the γ -ray energy spectrum consists of lines imbedded in a continuum. The lines are the characteristic emissions of the excited daughter species, while the continuum results from the Compton effect and bremsstrahlung.

6. Amersham-Searle ^{131}I - Stock # IBY.84, ^{85}SR - Stock # SOY.72, and ^{134}Cs - Stock # CCZ.72.

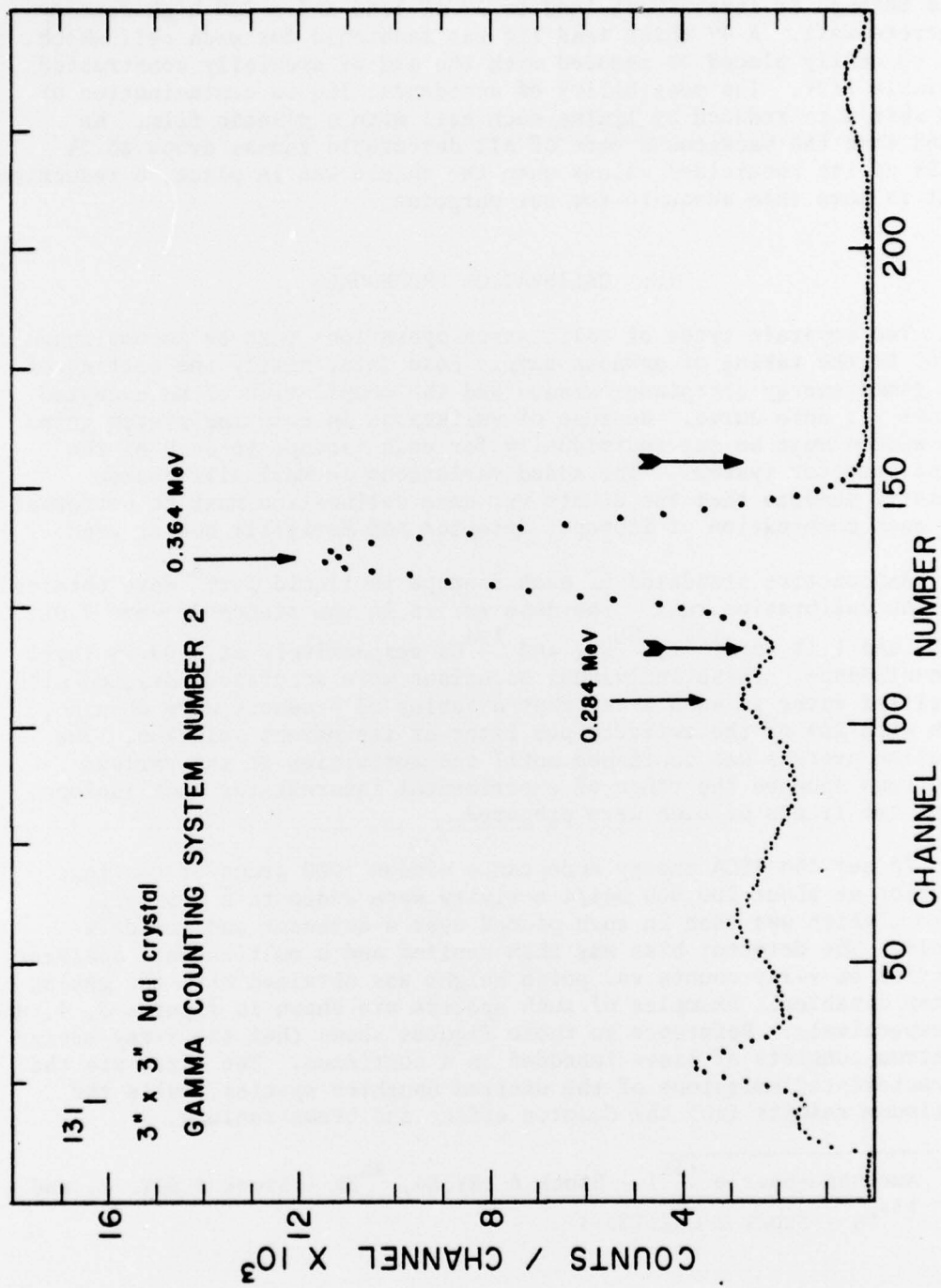


FIGURE 3. The gamma ray spectrum of ^{131}I .

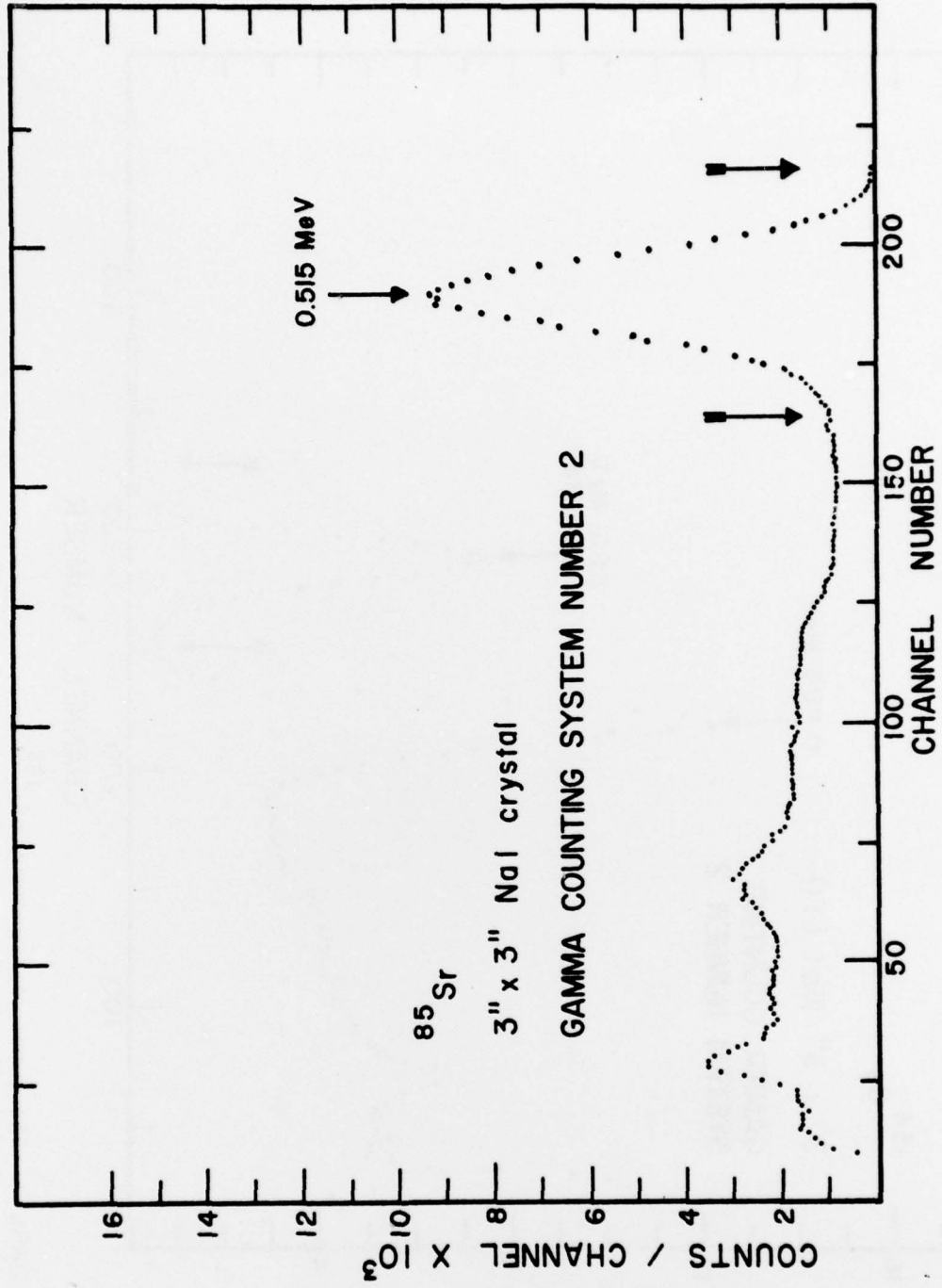


FIGURE 4. The gamma ray spectrum of ^{85}Sr .

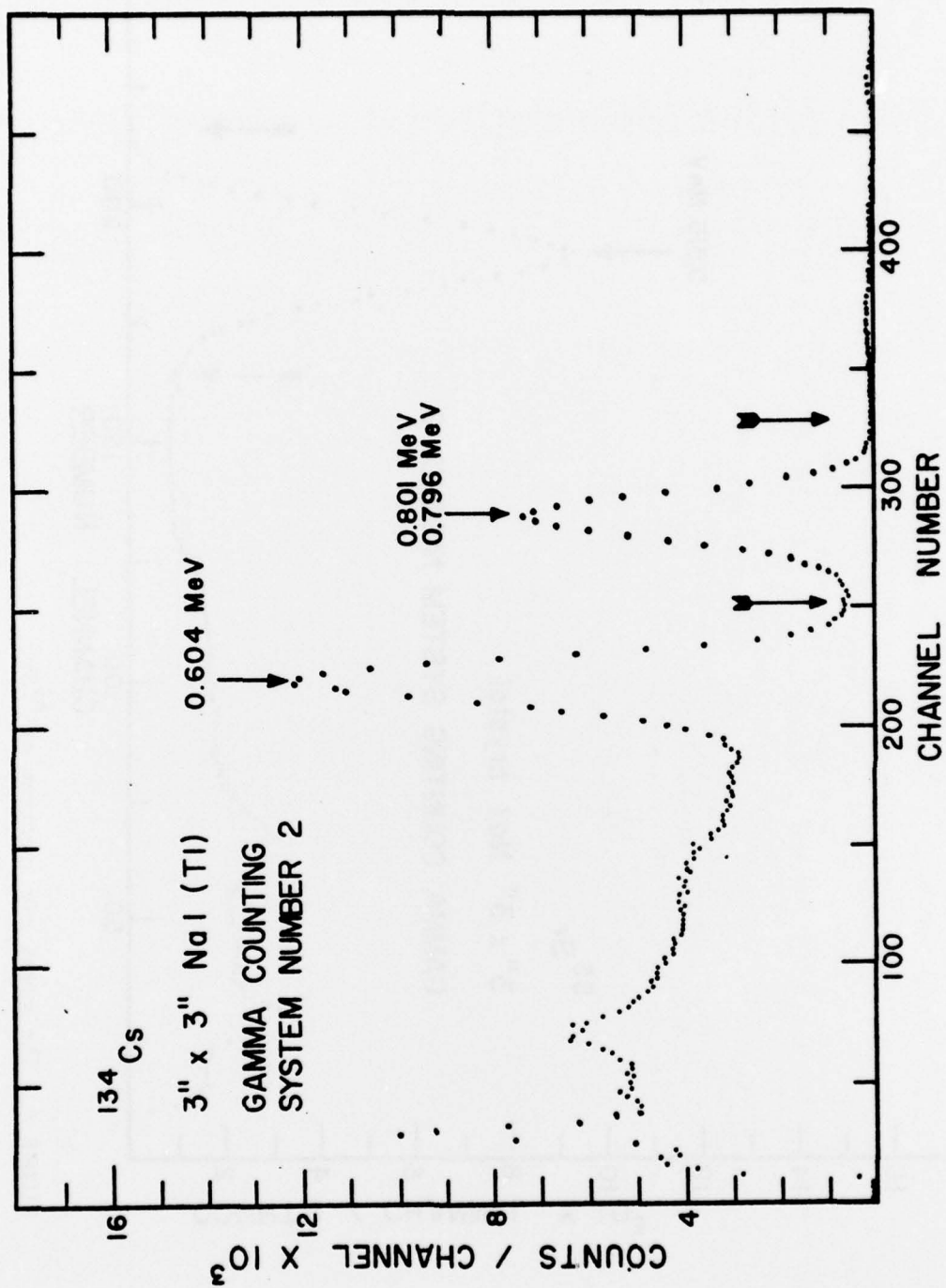


FIGURE 5. The gamma ray spectrum of ^{134}Cs .

Our experimental method consists of counting the unknown sample for a fixed time and ascertaining the net number of counts that fall in an energy interval surrounding one particular isolated, prominent line in the γ -ray spectrum. By comparing that rate with one determined in similar measurements upon the standards it is a simple matter to ascertain the dose. The energy limits are quite reproducible from day to day because the "valleys" on either side of the line provide ready points of reference. Setting the energy limits about a single line also assures us that the counts will be dominated by the characteristic emissions of the subject isotope and will not be heavily influenced by Compton scattered photons from shield components and other extraneous sources. After background was subtracted, no significant differences (be they due to pulse pile-up, detector gain changes, or amplifier drift) could be found between the gamma spectra obtained from either the standards or unknowns. This remained true over the entire activity range of interest.

To select only those γ -rays that fall into the energy interval E_1 to $E_1 + \Delta E$, estimates of the appropriate values of e_1 and Δe were set on the TSCA. The gate was then enabled and a spectrum of that portion of the gamma spectrum falling within the set limits was obtained and stored in a portion of the MCA memory adjacent to the ungated spectrum. The MCA has the capability of giving an overlapped display of portions of its memory. This allowed us to rapidly assess whether the set limits were the desired ones, and, by repeating the entire procedure a few times, zero in upon the correct settings. The interval settings were checked at least twice during each operating day. No variation was found during the entire course of the experiment. The limits used for each isotope are indicated by the tailed arrows in figs. 3-5.

Our next task was to determine a selected γ -ray counts vs. dose per liter curve. First a Marinelli beaker was selected, was weighed with a solution balance, and was loaded with exactly 1000 g of distilled water. The beaker was then placed over a particular detector inside a shield and the background activity in the selected energy interval was counted for exactly 30 minutes. Next, we replaced the distilled water with precisely 1000 g of the lowest dose standard solution and repeated the counting. The beaker was rinsed and the entire procedure was repeated with progressively more radioactive solutions. The net count due to each standard solution was determined by simple subtraction. The standard test dose $D_s(t)$ at the counting time t was ascertained by applying the usual exponential decay law

$$D_s(t) = D_s(T_0) \exp(-(t-T_0)/\tau) \quad (1)$$

to the dose $D_s(T_o)$, mean life τ , and measurement time T_o data provided by the standard's manufacturer as modified by our dilution procedure. The mean lives (not half-lives) of the three isotopes are ^{131}I -11.616 days, ^{85}Sr -93.36 days and ^{134}Cs -1085.7 days. The dose - counts curves for the three test isotopes ^{131}I , ^{85}Sr , and ^{134}Cs are shown in fig. 6.

The curves are fit by a linear dose-count relationship of the form:

$$D_s = \alpha(C-B) \quad (2)$$

where D_s is the standard dose in picocuries per liter, C is the raw gamma count, B is the background count, and α is the sensitivity constant. Values of α were determined for each combination of the three isotopes, three detectors and ten Marinelli beakers that were actually used in the tests by applying least squares methods to all the data points on each dose vs. count curve.

To measure the dose present in an unknown sample it is necessary only to determine its net count $(C-B)$ when that sample is counted utilizing the same sample preparation techniques and procedures as employed with the standards. Multiplication of that quantity by the appropriate value of α will then give the unknown sample dose, D'_x . ^{131}I and ^{85}Sr have lifetimes such that a significant portion of the activity might decay during RO operation, sample preparation, and counting procedures. For that reason the measured doses D'_x are corrected with the exponential decay law

$$D_x = D'_x \exp \frac{(t_x - T_B)}{\tau} \quad (3)$$

to compensate for decays in the time interval $t_x - T_B$ between an arbitrarily selected base time T_B (the instant the first sample of each run was taken) and the time the sample was counted t_x . The relative nature of the RO unit performance measurements permitted such arbitrary time base selection.

V. RESULTS AND ERRORS

During the fourteen running days of the R.O. test program more than 200 - ^{131}I , 150 - ^{134}Cs , and 100 - ^{85}Sr samples were counted. For the details of those results and the system's performance the reader is referred to the test report itself. The results are subject to

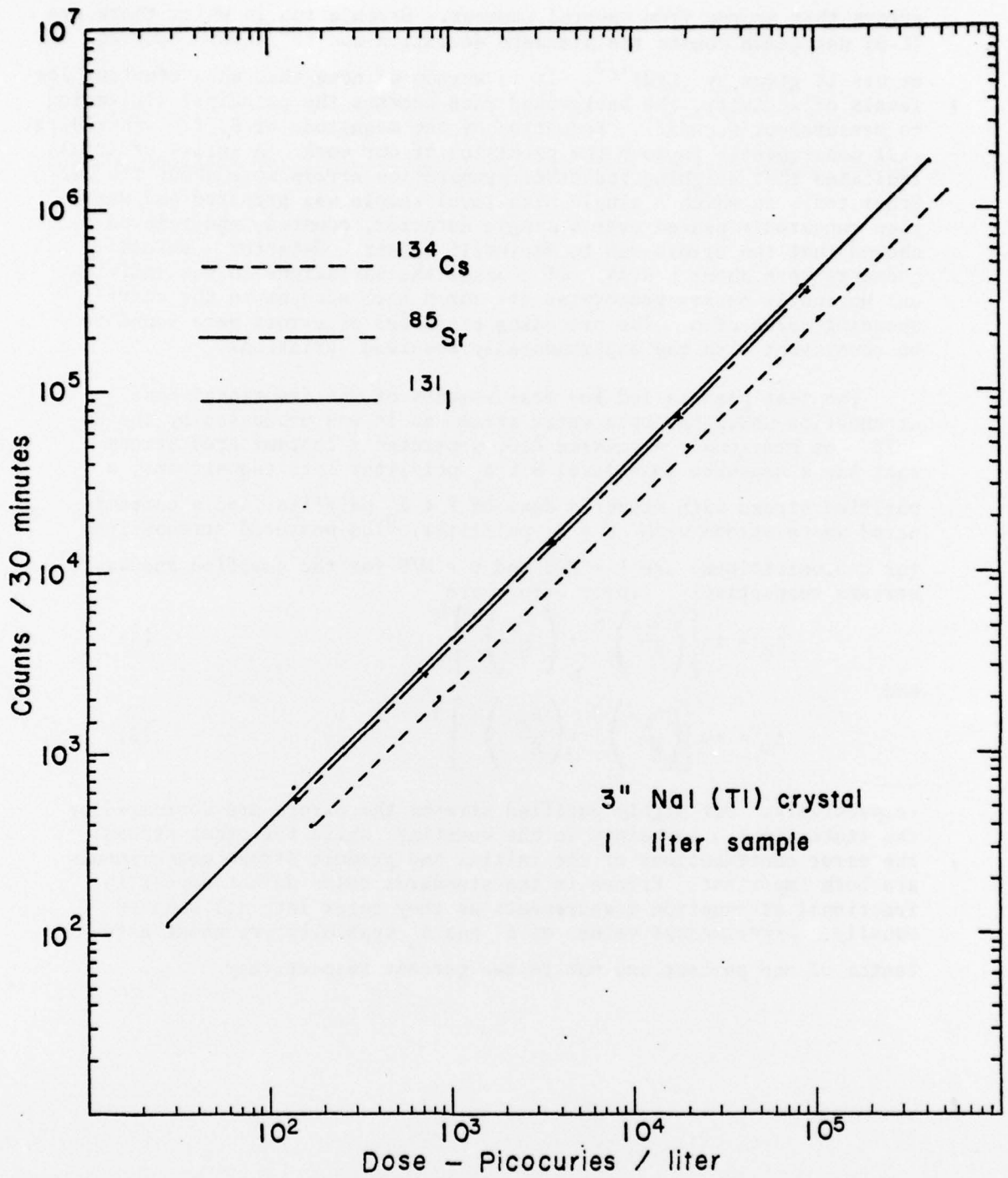


FIGURE 6. The dose-count curve for the three test isotopes.

errors that accrue from several sources. Given a run in which there are (C-B) net gamma counts the standard deviation due to random counting errors is given by $(C+B)^{1/2}$. It is worthy of note that when counting low levels of activity, the background rate becomes the principal limitation to measurement accuracy. Reduction of the magnitude of B, i.e. shielding, will consequently improve the precision of our work. A series of trials indicates that weighing and other preparation errors were about $\pm 0.2\%$. Other tests in which a single high level sample was prepared and was then repeatedly placed over a single detector, counted, and removed showed that the errors due to Marinelli beaker - detector - shield geometry were about $\pm 0.4\%$. Of course, the variations in the individual Marinelli beaker geometries are taken into account in the corresponding value of α . The preceding estimates of errors were found to be consistent with the experimentally observed variations.

The test plan called for measurements of the fractional dose attenuation undergone by a water stream as it was processed by the RO unit. As previously discussed R.O. separates a contaminated stream that has a measured dose level $S \pm \Delta_S$ pci/liter into two streams, a purified stream with measured dose of $P \pm \Delta_P$ pci/liter and a contaminated waste stream with $W \pm \Delta_W$ pci/liter. ^PThe measured attenuations (or concentrations) are $\pi = P/S$ and $\nu = W/S$ for the purified and waste streams respectively. Their errors are

$$\Delta_\pi = \pm \pi \left[\left(\frac{\Delta_P}{P} \right)^2 + \left(\frac{\Delta_S}{S} \right)^2 \right]^{1/2} \quad (4)$$

and

$$\Delta_\nu = \pm \nu \left[\left(\frac{\Delta_W}{W} \right)^2 + \left(\frac{\Delta_S}{S} \right)^2 \right]^{1/2} \quad (5)$$

respectively. For highly purified streams the errors are dominated by the statistical uncertainty in the counting, while for other streams the error contributions of the initial and product stream measurements are both important. Errors in the standards doses do not appear in fractional attenuation measurements as they enter into all samples equally. Experimental values of Δ_π and Δ_ν typically are about a few tenths of one percent and one to two percent respectively.

VI. SUMMARY

The counting system described in this report provides an efficient, simple and accurate means of counting gamma-emitting radioactivity in water samples. Each of the three counting systems that we constructed uses a 3 inch NaI(Tl) scintillator, a phototube, and associated electronics in combination with a lead/concrete shield. They are effective in the range between a few tens and several millions of picocuries/liter. Hundreds of measurements of the attenuation of activity that a stream undergoes during R.O. processing have been carried out. Each was done in a thirty minute counting time with an accuracy of a fraction of one percent. At levels above a few thousand picocuries/per liter the accuracy of the absolute activity measurements is limited by the quality of the calibration standards, while at lower levels statistical uncertainty is dominant.

ACKNOWLEDGEMENTS

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