INSTRUMENTAL METHOD FOR MEASUREMENT OF ^{131}I IN MILK

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SUMMARY

A prototype detector for radioiodine in milk has been tested using aqueous solutions of ^{131}I. Extrapolation of the test results indicate that a detector with a 0.25 liter sample volume can measure ^{131}I in milk to 0.5 picocuries per liter, with no chemical processing of the sample. Beta gamma coincidence counting keeps the background low. In this experiment we evaluated the counting efficiency, the accidental coincidence rate, and the cosmic ray coincidence background.

1. INTRODUCTION

This instrumental method is intended to replace radiochemical procedures for the analysis of ^{131}I in milk. To compete with these procedures (1) (2), we must identify the ^{131}I by its radiation instead of its chemical properties, reject other sources of radiation and achieve an efficiency for radioiodine so that a sensitivity of 0.5 pCil^{-1} is achieved. We use beta gamma coincidence counting to achieve these goals. All ^{131}I beta decays are accompanied by a suitable gamma photon, and 81% are in coincidence with the 364 KeV gamma (3).

2. DETECTOR

The short range of beta particles in milk makes efficient detection difficult. To overcome this difficulty, we use 167 rods of Pilot "B" plastic scintillator 0.25 cm diameter by 9 cm long, Fig. 1. These rods were mounted in a cup which held the sample. They were viewed at one end by a phototube. The volume of the cup, about 130 cc, was approximately equally divided between rods and sample. The cup, beta detector, and sample assembly was inserted into the 5.1 cm diameter x 7.6 cm deep well of a 12.7 diameter x 12.7 cm high NaI(Tl) gamma scintillation detector.

3. COUNTING SYSTEM

We assembled the beta gamma coincidence counting system from standard nuclear instrument modules. Important parameters were the beta and gamma energy windows, the coincidence (time) window, and the beta signal delay. The beta detector has a faster rising signal than the gamma detector. Therefore, we delayed the beta signal to maximize the coincidence rate, Fig. 2. To obtain maximum counting efficiency we had to set the energy windows to include some low energy noise. Thus, the gamma detector counted about 1.5 counts for each ^{131}I decay, and the beta detector counted about 2 counts for each decay. We were able to lower the threshold to a gamma rate of 14 and a beta rate of 70 counts per ^{131}I decay before any increase in the coincidence rate was seen. Setting the gamma window on the main peak gave an order of magnitude reduction in the single event rate at a cost of a factor of three in the coincidence rate. The same window on the beta channel cut the coincidence rate to 7% of the rate with the normal threshold. The coincidence (resolving time) window was normally 55 nsec. At 35 nsec and less, coincidence events were lost. Upper level discriminator signals from beta and gamma channels were cointed in coincidence to measure the cosmic ray event rate. Figure 3 shows the cosmic ray event rate as a function of energy. Extrapolating this rate to the ^{131}I window yields 15 cpm (MeV)$^{-1}$, or 9 cpm in the 600 KeV window.

4. RESULTS

Figure 4 shows the coincidence rate as a function of time. The disintigration rate of ^{131}I is plotted as a solid line. Coincidence counting efficiency, the ratio of the counts to the disintigrations, is 10%.

5. EXTRAPOLATION TO FULL SIZE DETECTOR

Our goal for the measurement of ^{131}I in milk is a lower limit of detection (LLD) of 0.5 $pCil^{-1}$. Pasternack and Harley (4)[For further discussion see Bowman and Swindle (5)] define LLD as:

$$LLD \simeq \gamma(k_\alpha + k_\beta)\ (C_{B+S} + C_B)^{1/2} \quad (1)$$

where:

γ converts units of Cs to units of LLD

$k_\alpha = 1.645$ (5% chance of false detection)

$k_\beta = 1.287$ (90% confidence of true detection)

C_{B+S} is the count during the measurement

C_B is the background count, measured for the same time

LLD = 0.5 pCi/l

γ is defined by the detector volume (V), counting time (T) and detection effeciency (ε):

$$\gamma = (pCi/DPM)(VT\varepsilon)^{-1} = (0.22\ VT)^{-1} \quad (2)$$

$$C_S = \frac{(LLD)}{\gamma} = 0.11\ VT \quad (3)$$

Therefore:

$$C_B/T \lesssim 1.41 \times 10^{-3}V^2T - 5.5 \times 10^{-2}V \quad (4)$$

Solving (4) with various values of VT yields Table I.

TABLE I

Parameters of detector to measure ^{131}I in milk at 0.5 $pCil^{-1}$

Active Volume liters	Counting Time Minutes	Background Rate CPM
0.25	1600	0.13
0.25	1300	0.10
0.25	720	0.05
0.50	1440	0.48
0.50	800	0.25
0.50	360	0.10

6. DISCUSSION

Realization of the detector described in Table I rests on three conditions:
(1) Maintain β efficiency .
(2) Maintain the same gamma efficiency while increasing the well volume to accomodate the larger detector.
(3) Keep the coincidence background rate to that specified by the detector volume and counting time.

Let us assess the chances of meeting these requirements.
(1) Our experience with scintillator rods indicates that light may be collected satisfactorily up to a length to diameter ratio of 100. To stay within this, we limit the length of the rods to 20 cm, while keeping the diameter the same (0.25 cm) as in the prototype.
(2) The NaI(Tl) well detector for gamma needs a well volume of 0.5 liter to accomodate an active volume of 0.25 liter. We obtain most of this by increased length. The diameter increased only 11% over the prototype. This represents only a 4% loss of ^{131}I photons (6).

(3) Lieshout, et al (7) discuss sources of background in large NaI(Tl) detectors. These may be separated into internal, various gamma and cosmic rays. Detection by the beta detector of a source internal to the gamma detector is very unlikely, and the internal background of the beta detector is very low. Penetration of both detectors by a gamma photon is unlikely, and the beta detector's efficiency for gamma is low. Thus, the principal coincidence background will be cosmic rays.

May and Marinelli (8) discuss the components of the cosmic ray background and their effect on the background of a low level scintillation detector. The ionizing component is readily detected in an anticoincidence guard. This is the major component, and in this laboratory we have achieved over 90% reduction in cosmic ray background with such a guard (9). The neutral component while penetrating the guard is unlikely to cause a coincidence count. However, a neutron may be captured in a plastic rod. The beta detector would detect such an event, and the resulting 2.2 MeV gamma photon has a high probability of being detected by the gamma detector. The upper energy cutoff, set at about 0.8 MeV for ^{131}I will reject most of the neutron capture gamma photons.

Thus to control background we have the techniques of coincidence, anticoincidence and energy resolutions. With these we expect to achieve a cosmic background event rate close to 0.1 CPM.

7. CONCLUSION

Our tests with a prototype β-γ coincidence detector for ^{131}I in milk indicate that a lower limit of detection of 0.5 $pCi l^{-1}$ is feasible. Replacing present chemical techniques with this instrumental method has significant economic advantages. The speed of this detector in measuring higher ^{131}I concentrations would be valuable in an emergency situation.

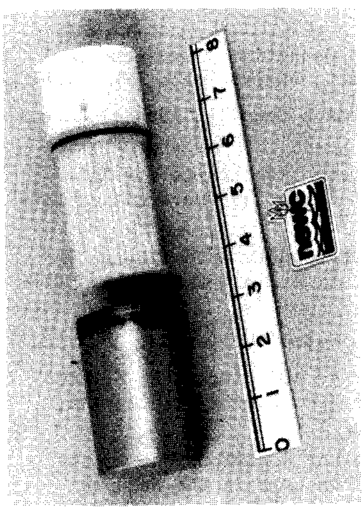

Figure 1
BETA DETECTOR

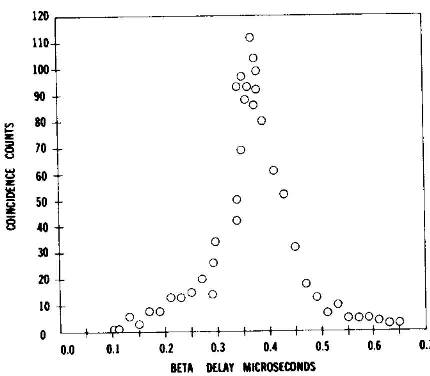

FIG. 2 COINCIDENCE COUNTING RATE V.S. BETA DELAY USING 35 NANOSECONDS RESOLVING TIME AND 0.5 MINUTE COUNTING TIME.

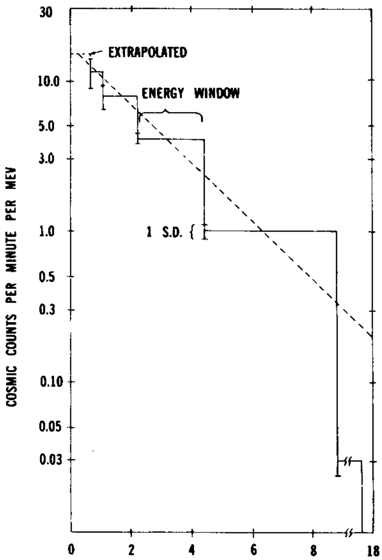

FIG. 3 COSMIC RAY COINCIDENT EVENT RATE V.S. UPPER LEVEL DISCRIMINATION (DIFFERENTIAL SPECTRUM DETERMINED FROM INTEGRAL DIFFERENCES)

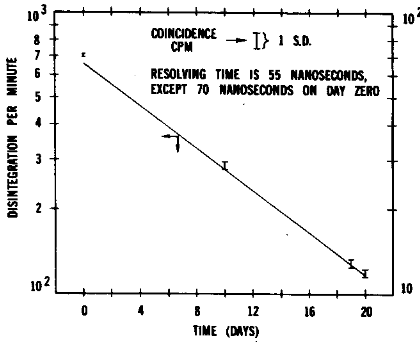

FIG. 4 IODINE 131 DECAY CURVE (LINE AND LEFT AXIS) AND COINCIDENCE CPM V.S. TIME (POINTS AND RIGHT AXIS)

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