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FISSION-PRODUCT MONITOR DEVELOPMENT

W. E. Draper

Nuclear Aerospace Research Facility
Fort Worth Division of General Dynamics
Fort Worth, Texas

Contract AF29(601)-6643

TECHNICAL REPORT NO. AFWL-TR-65-212
May 1966

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Research and Technology Division
Air Force Systems Command
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FOREWORD

This report was prepared by the Nuclear Aerospace Research Facility, Fort Worth Division of General Dynamics, Fort Worth, Texas, under Air Force Contract AF29(601)-6643. The research was performed under Program Element 6.54.02.12.4, Project 6773, Task 677304.

Inclusive dates of research were 1 October 1964 through 30 September 1965. The report was submitted 28 April 1966 by the AFWL Project Officer, Major Carl J. Schra (WLDN). The contractor's report number is FZK-269.

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This report has been reviewed and is approved.

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ABSTRACT

An investigation was conducted on improved methods of fission-product monitoring of the test reactors at the Nuclear Aerospace Research Facility (NARF), Fort Worth Division of General Dynamics. A literature review and the results of an experiment indicated the cascaded resin-bed system as the best method. In this system, a filter removes particulate matter, a cation bed removes most of the activity attributed to corrosion products, an anion bed removes fission products present in anionic form. In the case of a fission break, the principal activity collected in the anion bed results from the isotopes I-132 and I-134. A single-channel rate meter is employed to monitor a specific range of energies (820 $\pm$50 kev) corresponding to the most prominent gamma rays emitted by I-132 and I-134. Fission products can be identified by gross-gamma monitoring and their decay rates compared with the Way-Wigner equation. The delay in making this comparison is not in keeping with the fast response desired at NARF, and the system will not be sensitive to small pinhole leaks. A cladding failure of 1-mm^2 area for the Ground Test Reactor is postulated. Analytical results indicate that the activity attributed to the I-132 released from this area would be about equivalent to the background reading taken for a 162-Mw-hr run.

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SECTION I

INTRODUCTION

A reactor fission-product monitor (FPM) is a necessary adjunct to proper operation of the NARF Ground Test Reactor (GTR) and the Aerospace Systems Test Reactor (ASTR). The monitor should indicate the quantities and types of radionuclides that are being released. Compliance is assured by continuous monitoring of the primary coolants with detectors capable of discriminating against spurious radiation. This monitor should provide early warning of core failure to avoid (1) hazard to the public and operating crews, (2) serious damage to the reactor, and (3) expensive system decontamination.

1. Objective

This effort is directed toward the development of a fission-product monitor for the reactor cooling-water systems. The FPM should have a fast, reliable response and be suited to quantitative calibration with a minimum of effort. The monitors presently used at NARF do not meet these performance criteria completely.

2. Scope

A review of available literature was made to become acquainted with the methods of monitoring for fission products

at other facilities. Upon completion of the literature review the Reactivity Test Assembly (RTA) was used as the irradiation source to activate an aqueous solution of U-235. A closed-loop system consisting of ion-exchange resin beds and de-gassing facilities was used.

Detection efficiencies were determined for two methods of fission-product monitoring: gross gamma and resin-bed separation. Another method, monitoring beta activity of gases evolving from the irradiated solution, was also investigated. For this method, beta activity was below the threshold of detection.

SECTION II

LITERATURE REVIEW

Fission-product monitoring methods employed at several different facilities have been studied and their results compared to determine, economically and analytically, the most feasible mode for NARF to pursue.

1. Gas Analysis

The Savannah River Laboratory uses an automatic gas chromatograph to analyze the gases from primary coolant loops (Ref. 1). This method is not considered feasible for use at NARF because of calibration difficulties and the large amount of maintenance required. The time required for scanning a sample, on the order of ten minutes, is not compatible with NARF needs. A second system employed at Savannah River (Ref. 2) pumps gases evolving from the reactor primary coolant through a container of D_2O which is monitored by boron trifluoride (BF_3) neutron detectors. If fission-product gases are present, neutrons will be produced by the (γ,n) reaction in the D_2O and will be detected by the BF_3 counters. This method will not be considered because a quantitative calibration of this detector is difficult and the system is not as sensitive to small releases as other methods.

The electrostatic precipitation of positively charged fission-product gases is successfully employed at the EBR-I MARK IV (Ref. 3). This system, though ideally suited for use with sodium- or sodium-alloy-cooled reactors, would be difficult to adapt for use at NARF.

2. Airborne Activity

The Industrial Reactor Laboratories Reactor (Ref. 4) depends on the monitoring of airborne activities as a means of detecting fission-product release in the core. Detectors sampling air from near the pool surface, from the primary-system holdup tank, and from the facility exhaust system provide a sensitive method of detection. The integral nature of the GTR irradiation cell and the reactor, coupled with the wide variety of materials placed in the cell, would seriously restrict the effectiveness of a system of this type at NARF as it would require recalibration for each experiment.

3. Ion Exchange

The ion-exchange method discussed by Heath in Reference 5 is partially suited for use at NARF. In this system a portion of the primary coolant stream is diverted through a filter, a cation bed,and an anion bed in series. The filter, usually glass wool, removes solid particles from the water. The cation

bed will remove most of the corrosion products. Fission products are collected and monitored in the anion column. Monitoring is performed with a linear-amplifier system biased to exclude all pulses from gamma energies below 0.5 Mev. This method, with modifications, was investigated for use at NARF.

The modifications were to construct larger resin beds to obtain faster flow rates and quicker time response and to acquire the instrumentation necessary to monitor a specific range of energies for more positive identification of fission products.

SECTION III

EXPERIMENTAL EQUIPMENT

1. Irradiation Loop

With the resin-bed system, the method of calibration and detection, both quantitative and qualitative, would be to (1) dissolve a U-235 solution in water, (2) pass the solution through the Reactivity Test Assembly (RTA) for activation, and (3) analyze the fission products. A closed-loop system (Fig. 1) was constructed to accomplish these activities.

The holdup or de-gassing tank was constructed from a 2-ft section of 8-in. aluminum pipe. The upper end of the pipe was enclosed with a standard 8-in. flange and an 8-in. blind flange. A 1/2-in. hole in the blind flange allowed air to sweep over the solution. A well was built into the bottom of the tank to accommodate a 3- by 3-in. NaI(Tl) crystal.

The injection tank was constructed from a 5-gal glass jar with the bottom cut out. It was connected with tygon tubing to the pump (Little Giant Model 2).

A piece of 3/8-in. thin-wall aluminum tubing was inserted in a GTR rod element in the RTA core to serve as an irradiation loop.

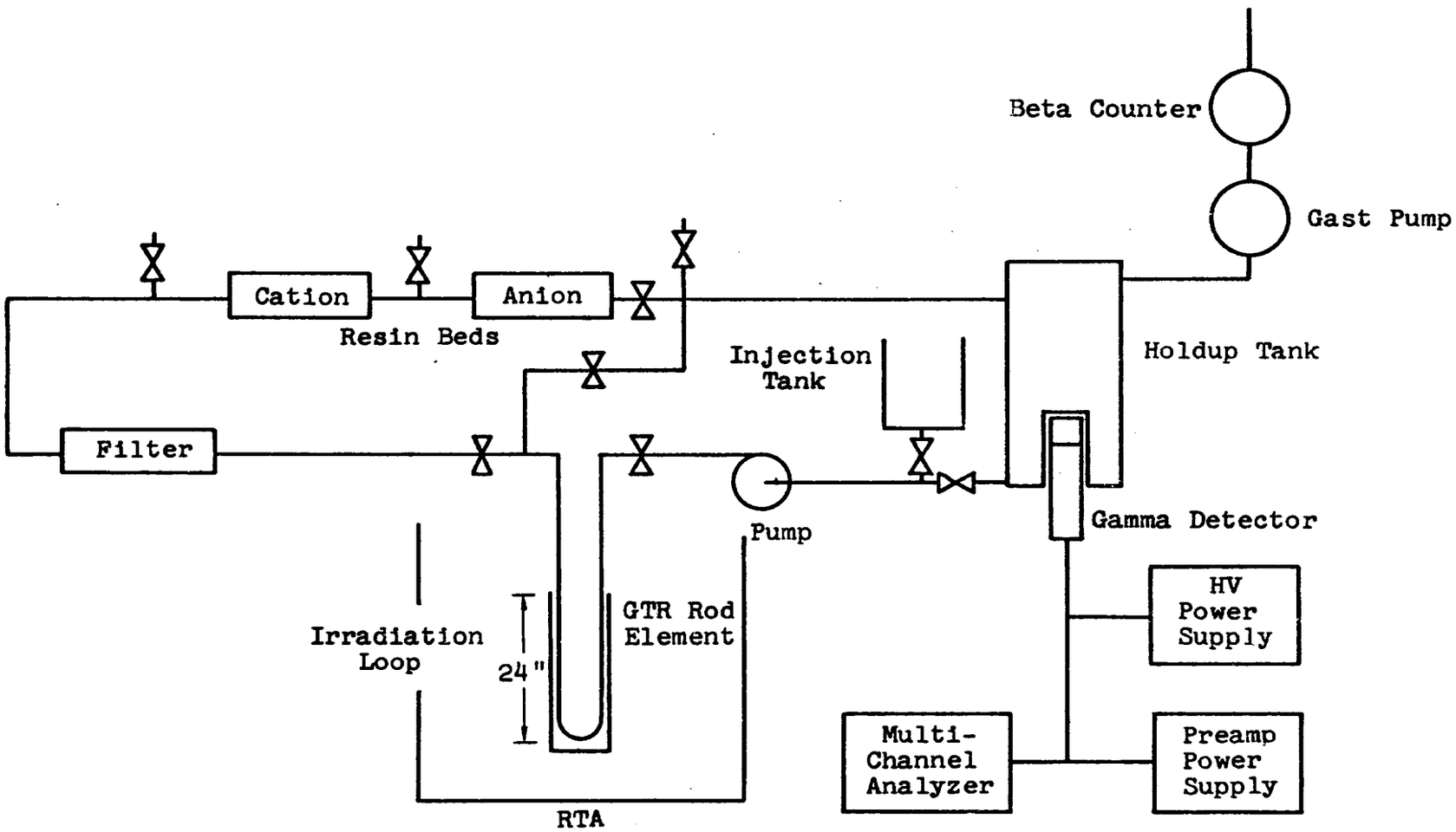


Figure 1 Drawing of the Experimental System

The filter and resin-bed containers, the cation and anion beds, were 4-in. sections of 2-in. stainless-steel tubing with standard pipe caps. The cation bed contained 70 cc of Amberlite IRA-400 and the anion bed contained 70 cc of Amberlite IR-120.

Screens were constructed from 40-mesh stainless-steel screen backed by 10-mesh screen for rigidity. The screens were held in place by snap rings seated in grooves machined on the inside of the 2-in. tubing. O-rings on either side of the screens prevented leakage of the resins.

A Gast Model 320 pump was employed to sweep air through the holding tank, together with any gases evolving from the irradiated solution, to an Atomic Accessories Model TSM 91 tritium counter for detection. Valves were arranged to permit bypassing the resin beds and to provide sample taps on either side of the resin beds.

A Fischer and Porter rotameter mounted at the holding tank was used to observe the flow. With this system a known concentration of U-235 could be added to the injection tank and, after exposure in the reactor, be passed through the resin beds or directly to the holdup tank.

2. Reactor Systems

The RTA is a low-power (less than 1000 watts), heterogeneous, light-water-moderated reactor using fully enriched MTR-type elements. It is especially designed to accommodate either a GTR or ASTR core mockup for studying various parameters affecting these cores. In addition, other experiments normally associated with a critical facility of this type are performed. The extreme flexibility of the facility and easy access to the core make it especially suitable as an activation source for the fission-product-monitor experiment. Figure 2 shows the RTA core with a GTR rod element in lattice position D-6.

3. Instrumentation

A 3- by 3-in. NaI(Tl) crystal (Harshaw type 12A 11.3) was used with a Dumont 6363 photomultiplier tube as the detector throughout the experiment. A Technical Measurements Corporation 400-channel analyzer was used to monitor the activity of the solution in the holdup tank. The resin beds were monitored with a TMC 100-channel analyzer because high background at the test facility necessitated removing the resin beds from the loop and transporting them to the Nuclear Laboratory.

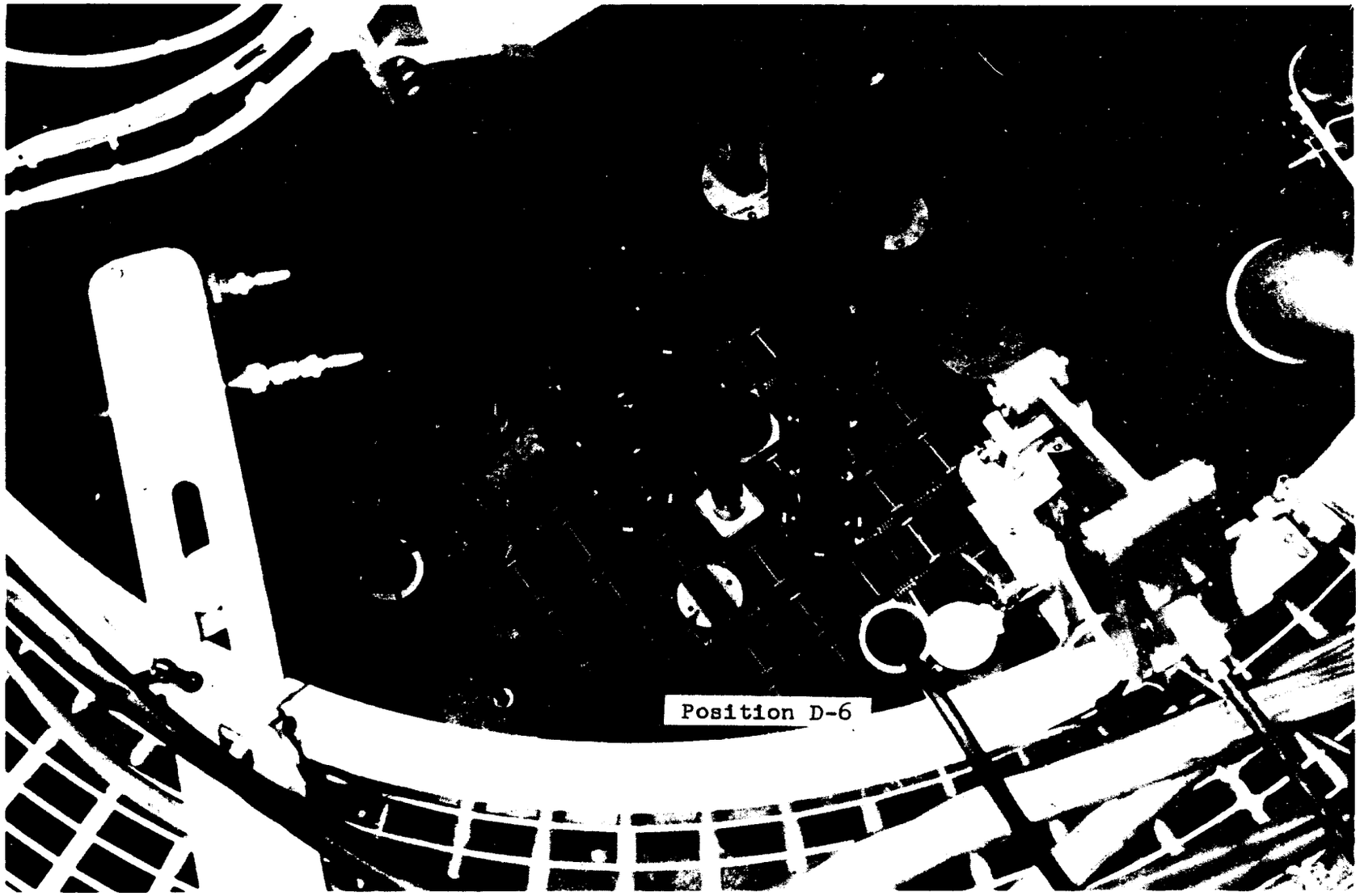


Figure 2 RTA Core with Rod Element in Position D-6

4. Fission Solution

The fission solution consisted of a 0.05-mℓ solution of uranyl nitrate, $UO_2(NO_3)_2$, containing 5.4 mg of U-235. This solution was then diluted with enough distilled water to make 10 mℓ of solution. The concentration of this solution in atoms/cc is calculated in the following manner:

$$\frac{5.4 \times 10^{-3} \text{ gm } U^{235}}{2.35 \times 10^{2} \frac{\text{gm } U^{235}}{\text{mole}}} \times \frac{6.023 \times 10^{23} \text{ atoms/mole}}{10 \text{ cc}}$$

$$= 1.38 \times 10^{18} \text{ atoms/cc}$$

Then, 2 cc of this solution were added to 10ℓ of distilled water, making a concentration of 2.76×10^{14} atoms/cc of U-235 in the solution to be activated.

SECTION IV

PROCEDURES AND DATA

1. RTA

a. Background Activity and Checks

The system was filled with distilled water for a background check. A typical background plot is shown in Figure 3. No well defined peak structure and a low count rate are the desired results, and this was obtained after a large shield consisting of lead and concrete blocks was constructed around the holdup tank. The RTA was then brought to power with the distilled water circulating to check for Na-24 and N-16. None was detected during this check.

b. Gross-Gamma Activity

The system was drained and the 10-ℓ solution with a concentration of 2.7×10^{14} atoms/cc of U-235 was added to the injection tank. The valves were set to bypass the resin beds and return the solution to the holdup tank after irradiation. The RTA was brought to power, and the flow was started and maintained at 100 cc/min. The reactor was shut down after all the solution had reached the holdup tank, where the solution decayed. A gamma spectrum was taken with the 400-channel analyzer at regular intervals. These data were examined for photopeak activity and decay characteristics.

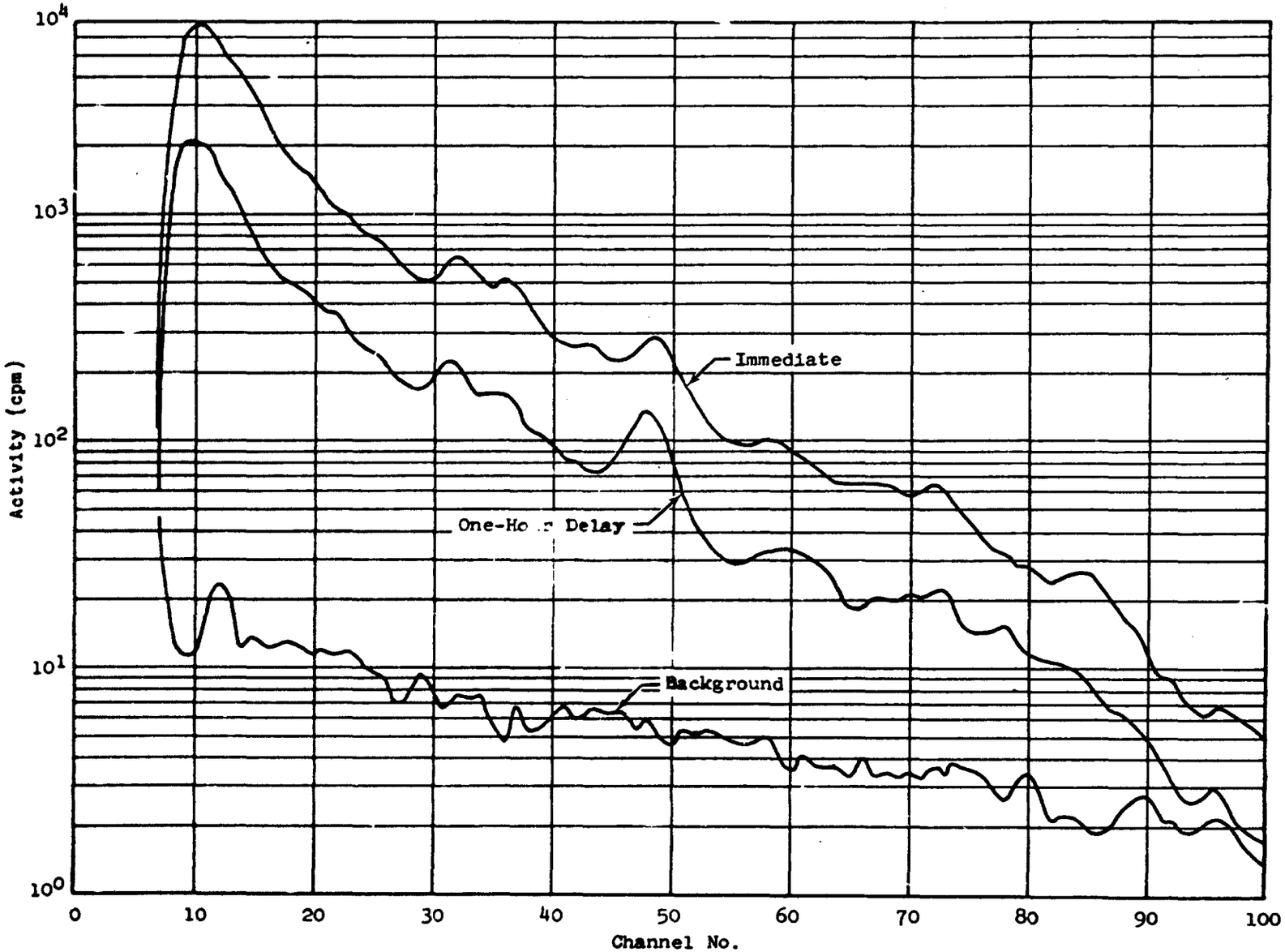


Figure 3 Gamma Spectrum of Activated Solution in the Holdup Tank

The gamma spectrum (Fig. 3) revealed broad complicated peak structure. Thus, in order to identify fission products as such, further separation was necessary. Figure 4 shows the similarity of the measured activity to the activity predicted with the Way-Wigner equation. A 10-min live time was used; therefore the first few points, in the region of greatest change, do not correlate as well because the solution was decaying rapidly as it was being counted. This does, however, tentatively identify the solution activity as being due to fission products. The delay in making this comparison, however, is not in keeping with the quick response desired in a fission-product monitor at NARF.

c. Beta Activity

While the gross-gamma activity was being investigated (see above), air was swept over the holdup tank and diverted over the beta counter. No activity was detected, and further investigation along this line was suspended.

d. Resin-Bed Activity

The same procedure used to measure gross-gamma activity was used to measure resin-bed activity, except that the solution was passed through the resin beds and, when the run was completed, the resin beds were removed from the system and taken to the Nuclear Laboratory for analysis with the 100-channel analyzer.

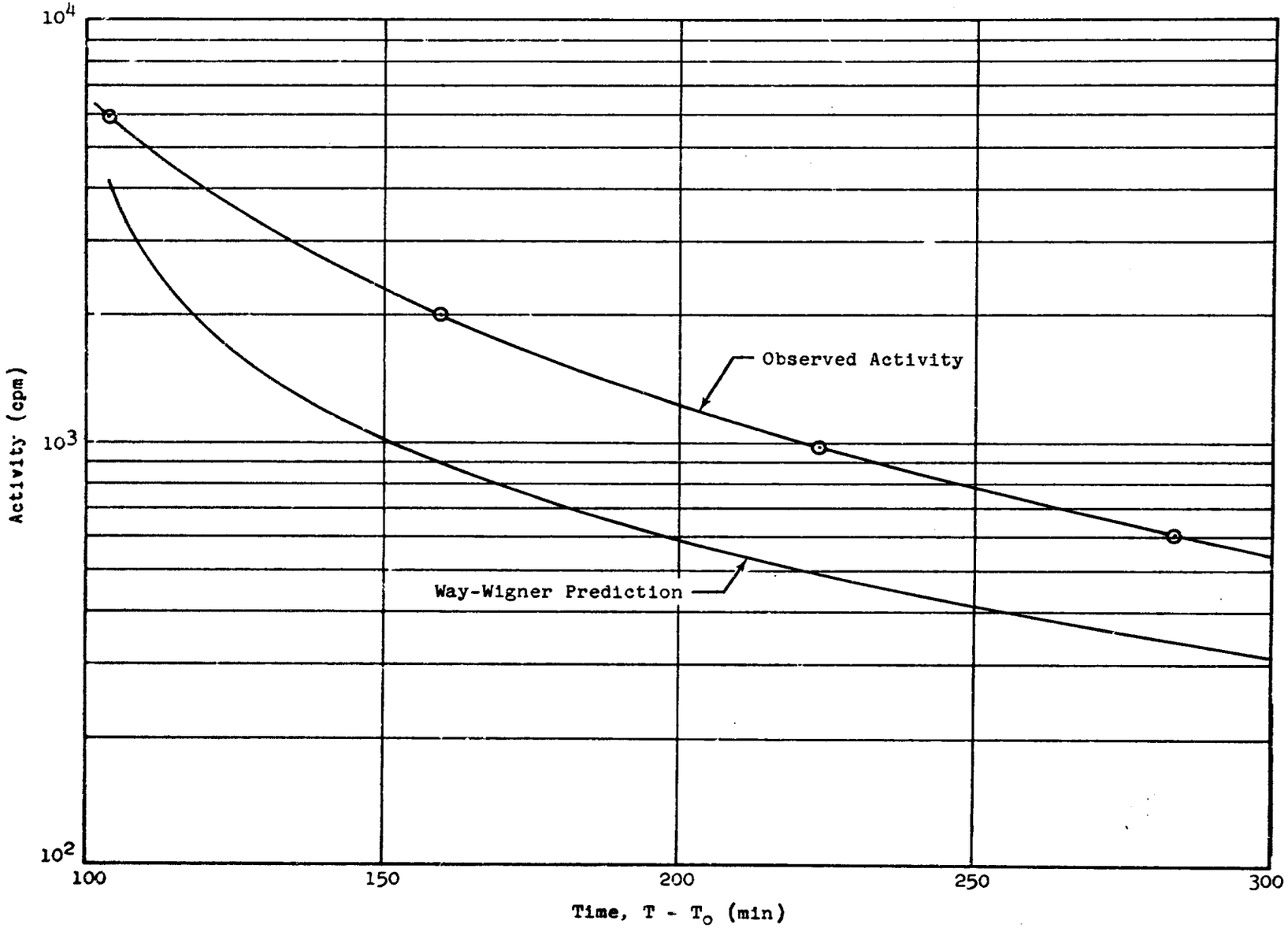


Figure 4 Measured Activity Compared to the Way-Wigner Equation

Figure 5 shows the gamma spectrum of the effluent collected on the anion bed. A strong peak is evident at 0.82 Mev; the decay of this peak corresponds closely to that of I-134. Later analysis of the decay of the anion bed showed the curve to be composed of at least two peaks that are identified as I-132 and I-134. The range of interest then falls about 50 kev on either side of the 0.82-Mev peak shown in Figure 5. This range (820 $\pm$ 50 kev) will include the 0.774-Mev gamma from I-132 and the 0.848-Mev gamma from I-134. These are the major contributors (as will be shown later) from the anion bed.

The fission-product activity collected on the cation is shown in Figure 6. These data are of academic interest only, since the corrosion products that will be collected by this bed in the GTR and ASTR systems would mask out the fission-product spectrum. It is of interest, however, to note that the predominant peak falls at 1.3 Mev, which, in the case of the GTR and the ASTR, would not be discernible because of the large amount of Na-24 (1.37 Mev) in the systems.

2. GTR

After the RTA tests were completed, an identical-type anion bed was installed in the existing GTR FPM loop. Figure 7 shows the activity collected on the anion bed taken from the GTR after 162 Mw-hr of operation. The absence of any peak at the range of interest (820 $\pm$ 50 kev) is important in that it

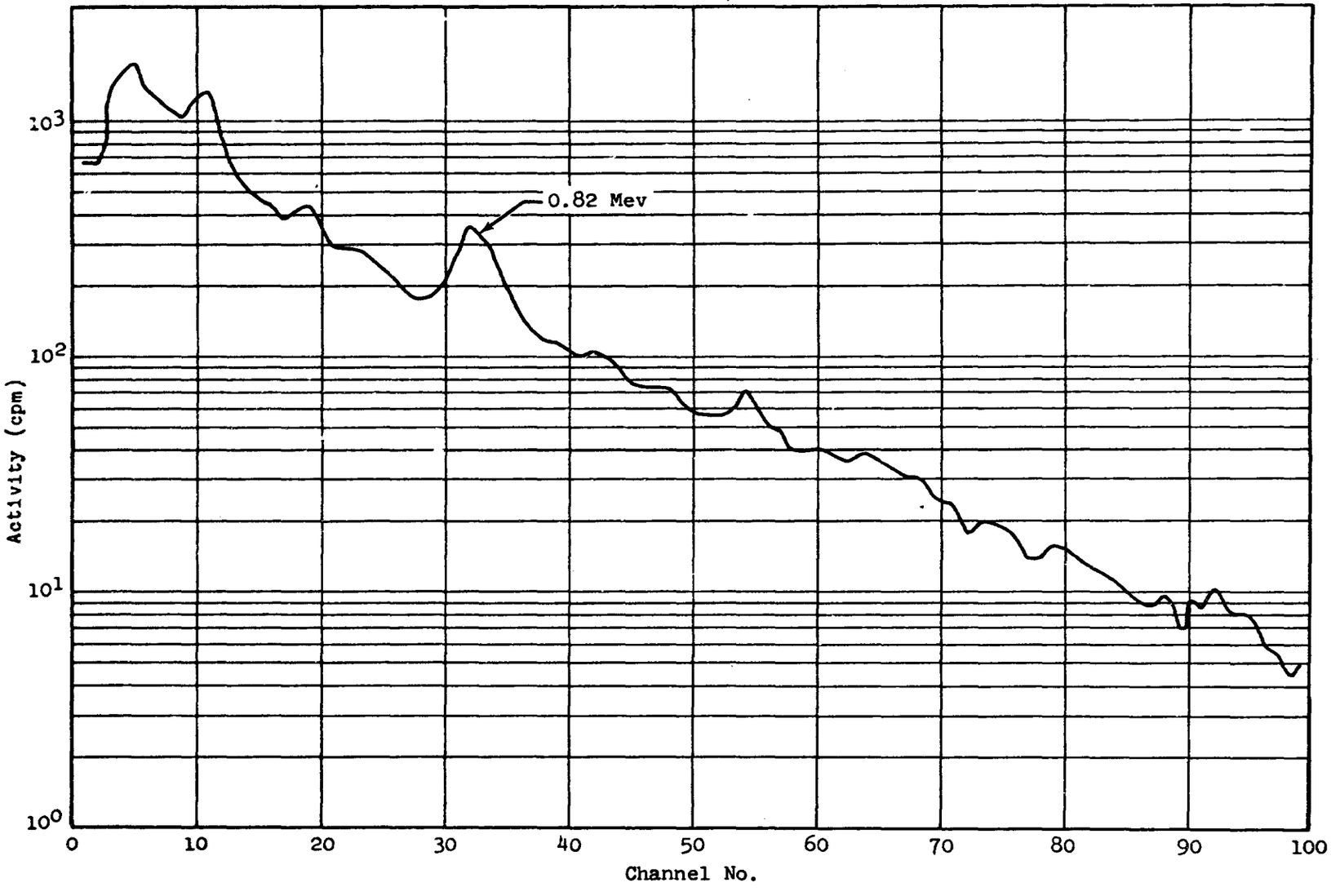


Figure 5 Spectrum of Gamma Rays from Anion Bed

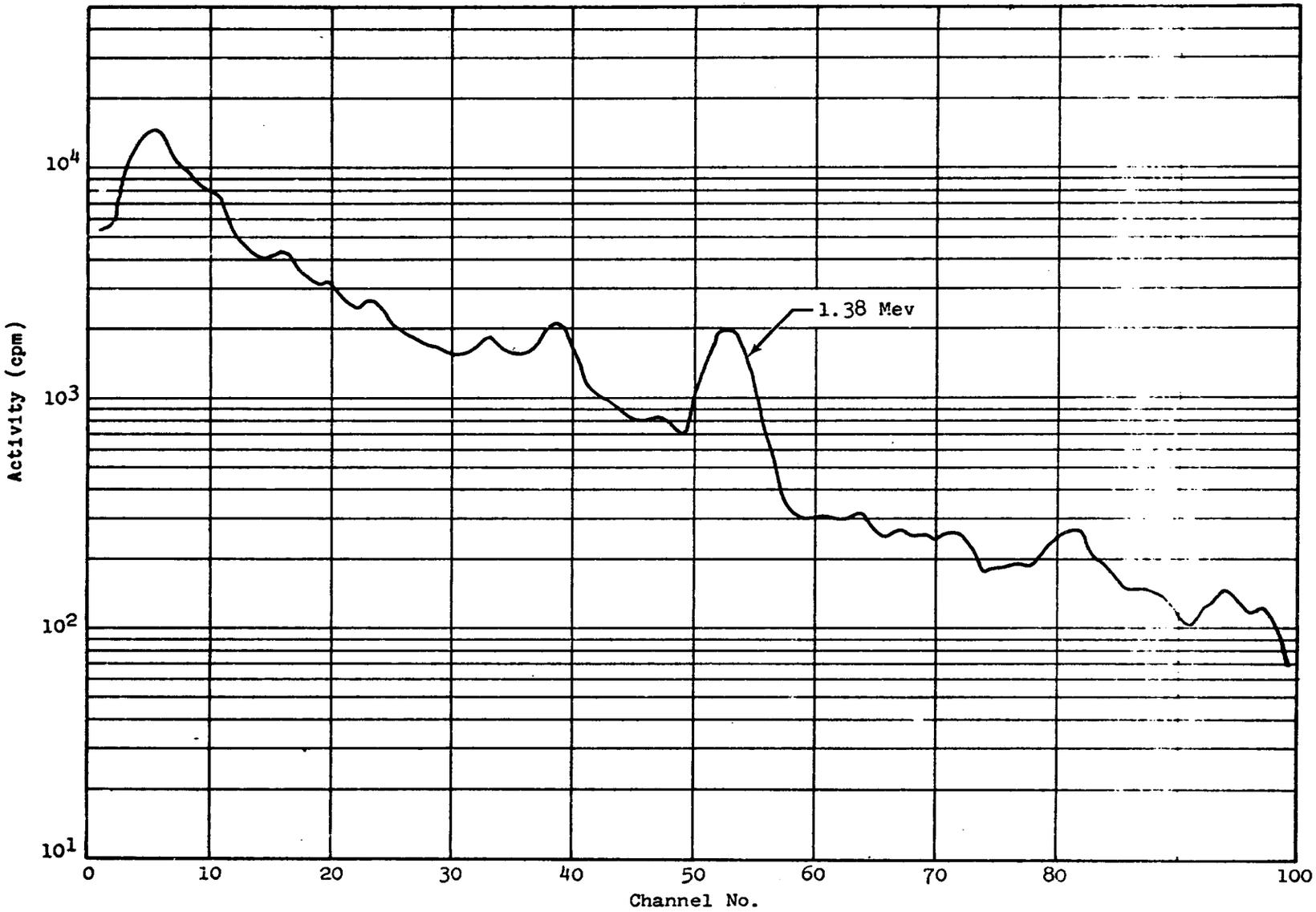


Figure 6 Spectrum of Gamma Rays from Cation Bed

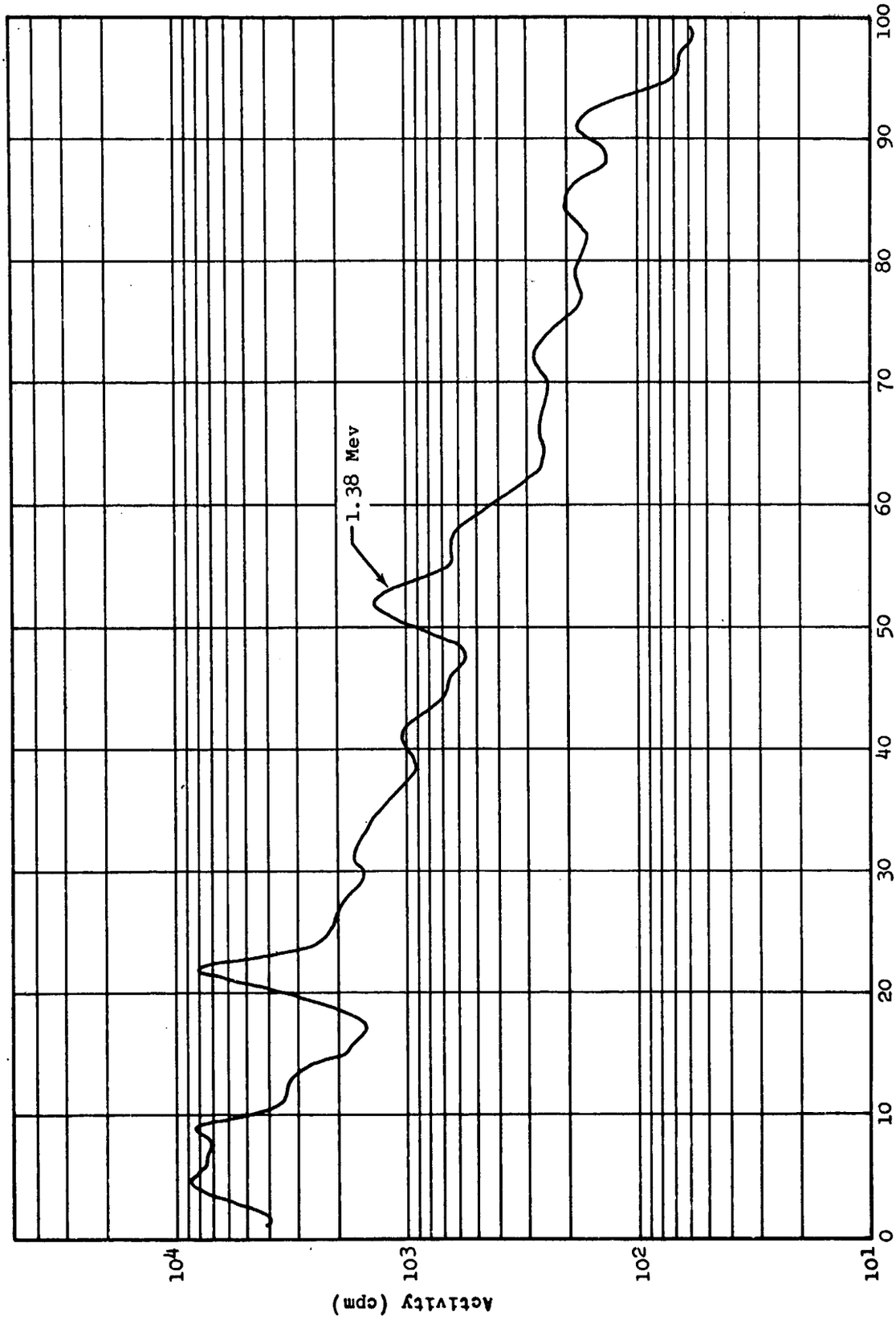


Figure 7 Spectrum of Gamma Rays from Anion Bed after 162 Mw-Hr of Operation

indicates that there are no leaks in the GTR core and that photopeak counting would be an effective means of identifying a fission break if it did occur. Another important fact is that only about 1000 counts/min are indicated in the range of interest. This leads to the assumption that rather small pin-hole leaks could be readily detected (see Section V.3).

SECTION V

ANALYSIS

1. Fission Rate in the Test Loop

The fission rate in the test loop can be calculated in the following manner (Ref. 6):

$$F = \bar{\phi}\sigma_f U^{235} q \qquad (1)$$

where,

$\bar{\phi}$ = the average thermal-neutron flux = 3.4×10^9 n/cm^2-sec

σ_f = fission cross section of U^{235} = 550×10^{-24} cm^2 (Ref. 6, p. 109)

U^{235} = concentration of U^{235} = 2.76×10^{14} atoms/cm^3

q = volume of the solution in the core = 62.8 cm^3

Thus,

$$F = (3.4 \times 10^9)(550 \times 10^{-24})(2.76 \times 10^{14})(62.8)$$

$$= 3.24 \times 10^4 \text{ fissions/sec}$$

2. Holdup-Tank Activity

The activity in the holdup tank can be calculated using the fission rate (above) and the equation (Ref. 6, p. 120)

$$A = 1.4P\left[(T - T_o)^{-0.2} - T^{-0.2}\right] \qquad (2)$$

where

A = expected activity, curies

P = power, watts

T = startup to count time, days

T_o = exposure time, days

The power, P, is $\frac{3.24 \times 10^4 \text{ fissions/sec}}{3.1 \times 10^{10} \text{ fissions/sec-watt}}$ or 1.04×10^{-6} watts. For every curie there is an equal amount of betas and gammas, so Equation 2 is altered by dividing the constant 1.4 by 2 (Ref. 7, p. 102). The equation then becomes

$$A = 0.70P \left[(T - T_o)^{-0.2} - T^{-0.2}\right] \text{ curies} \qquad (3)$$

Solving for $T = 104$ min, $T_o = 100$ min,

$$A = (0.70)(1.04 \times 10^{-6})(3.24 - 1.69) = 1.128 \times 10^{-6}$$

curie, or 3.7×10^{10} dps/curie $\times$ 1.128×10^{-6} curie $= 4.17 \times 10^4$ dps as the total activity in the holdup tank at $T - T_o = 4$ min.

The count rate recorded by the detector in the holdup-tank well was 5.8×10^4 counts/min at $T - T_o = 4$ min. The predicted count rate, derived by the procedure discussed in Appendix A, is 6.0×10^4 counts/min.

3. Resin-Bed Activity

The area under the total absorption peaks in the channel interval 28 to 38 (Fig. 5) divided by the accumulation time yields a count rate of 850 counts/min for the related gamma rays. The activity under the peaks is attributed to a combination of several isotopes of iodine. A list of these isotopes and their percentage gamma contribution in the range of

interest (820 ± 50 kev) is given in Table I (Ref. 8). If it is assumed that all of the iodine isotopes are collected on the anion bed, then the disintegration rate on the bed from each isotope, i, as a function of time may be calculated as follows:

$$A(T)_i = FP_i\gamma_i \left[e^{-\lambda_i(t_2 - t_1)} - e^{-\lambda_i t_2} \right] \qquad (4)$$

where

$A(T)_i$ = disintegration rate for each iodine isotope, dps

F = fission rate = 3.24×10^4 fissions/sec

P_i = probability that a gamma will occur in the range of interest

γ_i = fission yield

λ_i = decay constant, sec^{-1}

The derivation of Equation 4 is given in Appendix B. The disintegration rates were calculated for each isotope and are listed in Table II, where it can be seen that 2.3-hr I-132 and 53-min I-134 are the major contributors to the activity collected. Comparing the measured count rate with the sum of the I-132 and I-134 calculated count rates, we get $850/(2.28 \times 10^4)$, or a factor of 3.72×10^{-2}. This factor is not to be construed as a counting efficiency but is rather

Table I

IODINE ISOTOPES OF INTEREST

Isotope	Half-Life	Decay Constant (sec^{-1})	Fission Yield (Fractional)	Gammas at 820 ± 50 kev (%)
I-130	12.6 hr	.1528(-4)	.5622(-4)	70
I-131	8.05 d	.9965(-6)	.9454(-4)	2.8
I-132	2.30 hr	.8371(-4)	.1670(-2)	88.5
I-133	20.8 hr	.9256(-5)	.6552(-2)	9
I-134	53 min	.2179(-3)	.2170(-1)	100
I-135	6.68 hr	.2882(-4)	.2851(-1)	0
I-136	84 sec	.8251(-2)	.3924(-1)	3

Table II

CALCULATED ACTIVITY FROM IODINE ISOTOPES

(dpm at 820 ± 50 kev)

Isotope	Activity at $t_2 - t_1 = 10$ min
I-130	17
I-131	0
I-132	8.20×10^2
I-133	49
I-134	2.19×10^4
I-135	0
I-136	16

a composite of several factors, e.g., plate out, uptake efficiency of the anion bed, shielding from the walls of the bed container, and efficiency of the counting system in general.

The activity resulting from a 1-mm^2 cladding rupture postulated for the GTR and using the factor above would be calculated in the following manner, assuming an instantaneous release of all the iodine from the 1-mm^2 hole in a fuel plate and assuming a homogeneous mixture of this release in the GTR pool after a 100-hr run at 3 Mw: Iodine-132 for all fuel elements in the GTR core is

$$I^{132} = \frac{\gamma \Sigma_f \bar{\phi} N}{\lambda} \qquad (5)$$

where

γ = fission yield = 0.167×10^{-2}

Σ_f = U^{235} fission cross section = 5.5×10^{-22} cm^2

$\bar{\phi}$ = average thermal neutron flux in GTR at 3 Mw

= 1.6×10^{13} n/sec-cm^2

N = number of atoms of U^{235} in the core

$$= \frac{4.7 \times 10^3}{2.35 \times 10^2} \times 6.02 \times 10^{23} = 1.20 \times 10^{25}$$

Then,

$$I^{132} = \frac{\gamma}{\lambda} (1.6 \times 10^{13})(5.5 \times 10^{-22})(1.20 \times 10^{25})$$

$$= \frac{\gamma}{\lambda} (1.05 \times 10^{17})$$

The activity in dps is λI^{132}, which gives an activity of $\gamma(1.05 \times 10^{17}) = 0.167 \times 10^{-2}\ (1.05 \times 10^{17}) = 0.175 \times 10^{15}$ dps, the total activity in the core due to I^{132}.

The volume of the GTR pool is 7.56×10^{7} cm^3 and the surface area of all fuel elements in the core is 3.76×10^{5} cm^2. The activity of the pool water due to a 1-mm^2 hole in one fuel element is then

$$\frac{(0.175 \times 10^{15} \text{dps})\ (1\ mm^2)}{(3.76 \times 10^{7}\ mm^2)(7.56 \times 10^{7}\ cm^3)} = 0.0616\ \text{dps/cm}^3$$

or 3.7 dpm/cm^3. A 10-ℓ sample as used in the RTA experiment would yield 37×10^{3} dpm. This, times the efficiency factor, would yield $37 \times 10^{3} \times 3.72 \times 10^{-2} = 1.37 \times 10^{3}$ counts/min. Figure 7 shows the gamma spectrum of an anion bed taken after 162 Mw-hr of operation, where about 10^{3} counts per minute are indicated at the energy of interest. A count rate from the iodine of 3 times background should be sufficient for easy detection; therefore, a 3-mm^2 hole in a fuel plate should be easily detected.

SECTION VI

CONCLUSIONS AND RECOMMENDATIONS

The major conclusions and recommendations concerning the fission-product monitoring investigation are as follows:

1. Gross-Gamma Method

This method would not be sensitive to small pinhole leaks, and a delay-time would be required for the identification of any fission products that did exist.

2. Resin-Bed Method

The cascaded resin beds (ion-exchange columns) provide an effective method for separating fission products from other sources of activity. The experimental results indicate a rapid response to fission-product releases, and it has been postulated that small pinhole leaks on the order of 3 mm^2 of fuel surface could be detected.

3. New Equipment

Figure 8 is a diagram of new equipment that will be installed for use at NARF. The resin beds and filters will be the same as those used in the experiment. For routine monitoring, a single-channel analyzer and a rate meter with alarm provisions will be used. The window on the analyzer will be adjusted to cover the range of interest, i.e., 820 $\pm$ 50 kev. During normal operation, an increasing count rate,

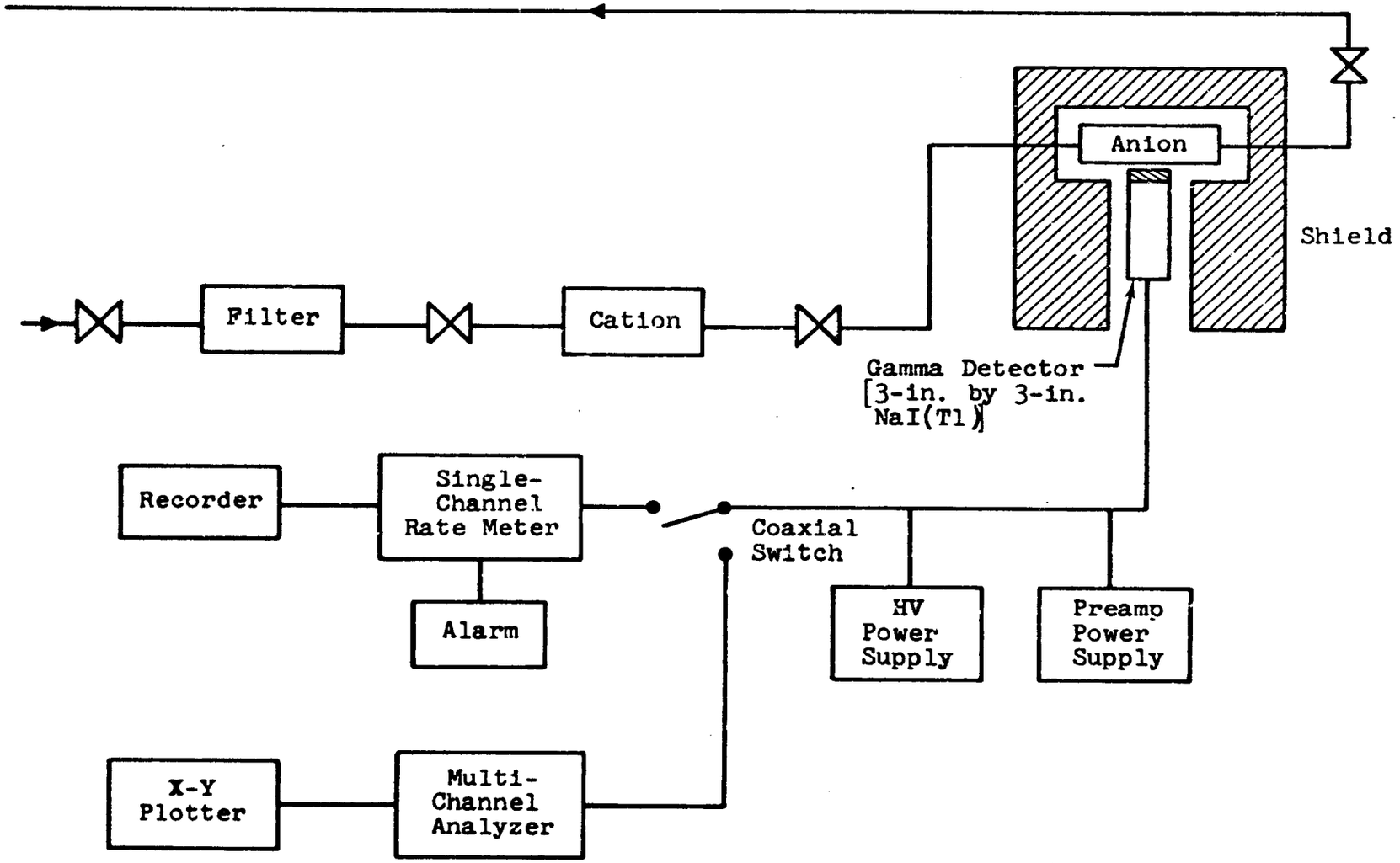


Figure 8 Diagram of New Equipment Installation

over previously established norms, would indicate the presence of fission products.

A 100-channel analyzer and a point plotter have been procured to analyze the activity collected on the anion bed and establish a normal curve for all operating conditions. Thus, in event of an alarm condition, a quick reference can be made to establish the cause.

4. Dynamic Tests

The experimental results have shown that the resin-bed system is sensitive to small pinhole leaks and, therefore, should be sensitive to large bursts as well. These results were obtained under controlled conditions, so that the operator knew what to expect. Conclusions concerned with the effectiveness of the system under actual operating conditions will have to await the performance of a dynamic test. Since all reactor systems are checked, it is recommended that a carefully controlled release of fission products be made in the GTR system.

APPENDIX A

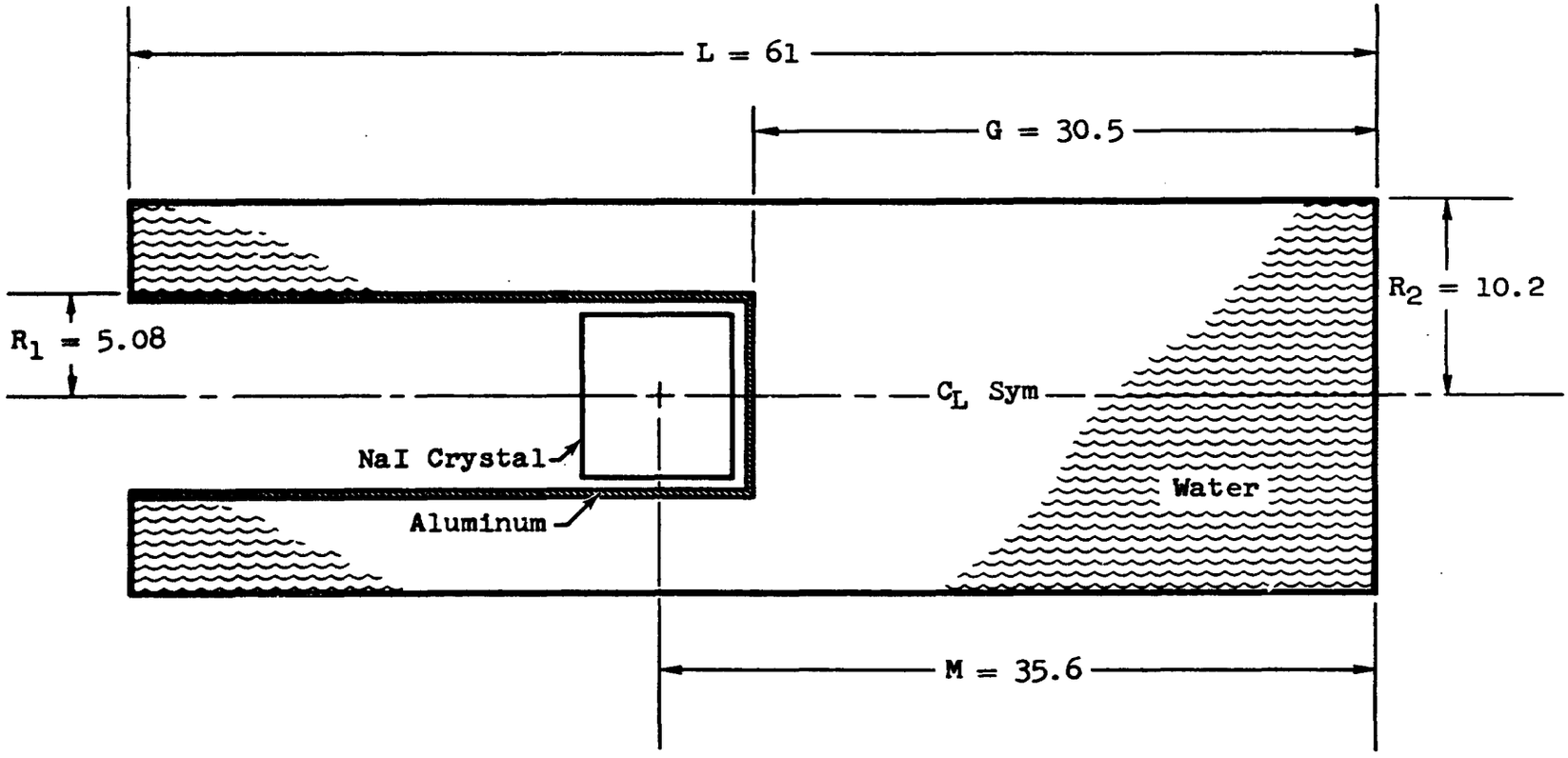
CALCULATIONAL PROCEDURES FOR GAMMA-RAY SPECTROMETER RESPONSE

The calculational procedures, and assumptions thereto, designed to predict the response of the gamma-ray spectrometer to decay gammas following insertion of radioactive products into the water surrounding the crystal are described.

The actual experimental geometry is shown in Figure A-1. Contaminated water is inside a 20.3-cm-ID by 61-cm-long pipe. A 3-in.-diameter by 3-in.-long NaI detector is in a 10.2-cm-OD by 30.5-cm-long aluminum tube with a 0.16-cm wall thickness and positioned as shown. Not shown in the figure is the thickness of MgO reflective coating, housing, and photomultiplier assembly of the NaI crystal. To simplify calculations, an assumption is made that the crystal is a sphere, of radius r_o, whose volume is equivalent to the actual volume of the crystal, or

$$r_o = \left(\frac{3}{4} r_n^2 L_n \right)^{1/3} = 4.35 \text{ cm} \qquad \text{(A-1)}$$

where r_n and L_n are the radius and length of the crystal, respectively.



Note: All dimensions in cm

Figure A-1 Experimental Geometry for NaI Gamma-Ray Spectrometer

The revised geometry, using r_o , for calculating the count rate C_γ of the NaI detector is shown in Figure A-2, where the thickness T of aluminum surrounding the crystal denotes an average thickness that includes detector housing, aluminum pipe, and crystal reflective materials. The value assumed for T is 0.6 cm.

The gamma rays emitted at the point isotropic source $S_o(t)$, shown in Figure A-2, are polyenergetic. In addition, the detector count rate will include gammas that have scattered in the media surrounding the NaI crystal, primarily Compton scattering. It will be assumed that the average energy of the gamma ray incident on the crystal is 0.7-Mev. A buildup factor will be introduced to account for gamma rays scattering prior to entering the crystal.

Consider a time-dependent isotropic source of monoenergetic gamma rays $\Gamma S_o(t)$ in the volume element dV whose spherical coordinates are ℓ, θ, and ρ . $S_o(t)$ is in units of disintegration per unit time and Γ denotes the number of gamma rays produced per disintegration. The probability that a gamma ray in dV will be emitted in the appropriate direction so as to pass through a volume element dV_N in the NaI crystal is

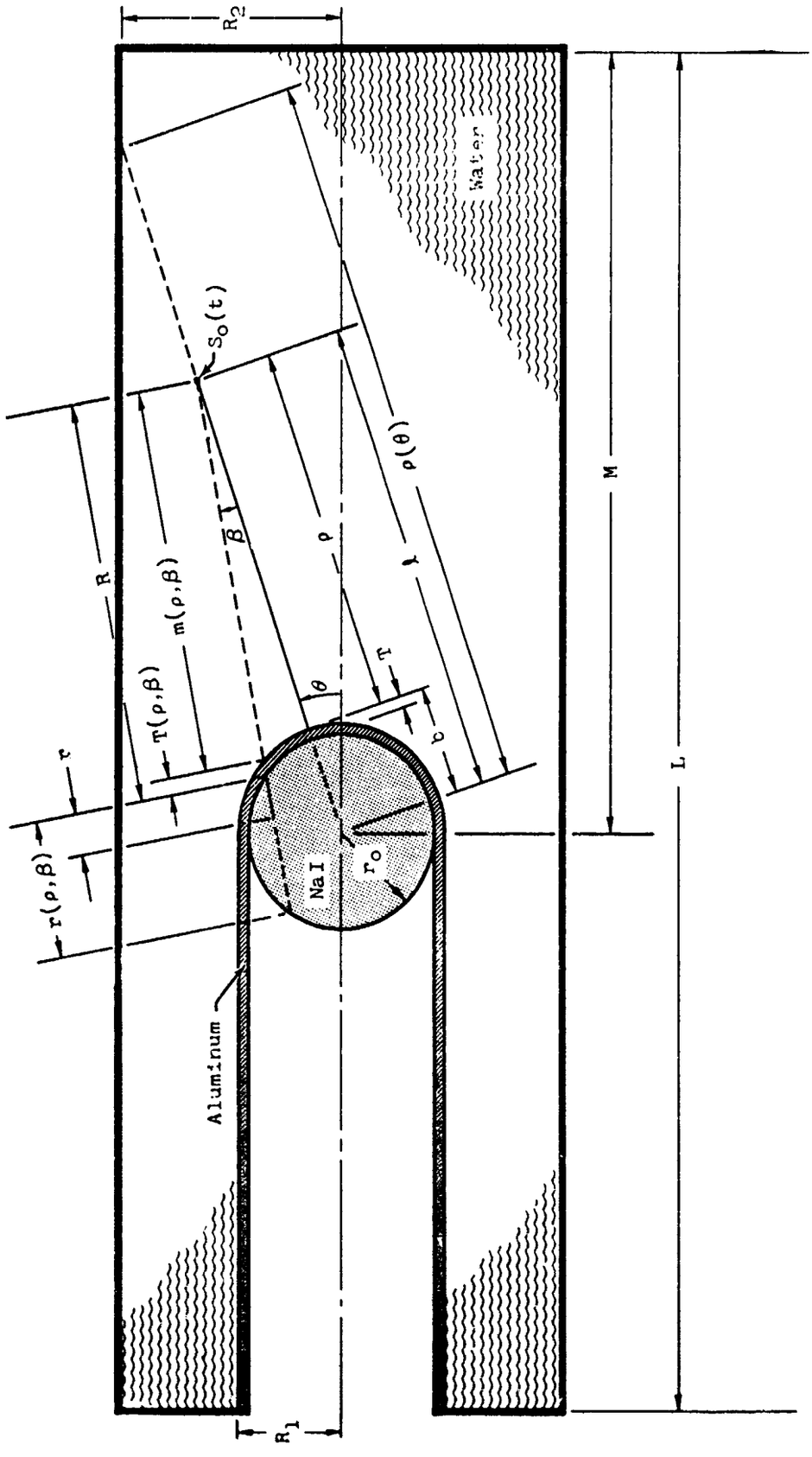


Figure A-2 Calculational Geometry

given by the fractional solid angle subtended by dV_N at the emission point, which is $\frac{1}{4\pi R^2} R^2 \sin\beta \, d\gamma \, d\beta$. The probability that gamma rays which have their direction of motion within this solid angle will reach the volume element dV_N is $e^{-\mu_1 m(\ell,\beta)} e^{-\mu_2 T(\ell,\beta)} e^{-\mu_3 r}$, with μ_1, μ_2, and μ_3 being the linear total attenuation coefficients for water, aluminum, and NaI, respectively. The probability that a gamma ray which reaches dV will undergo a reaction to produce a count is $\mu_3 \, dR$.

The count rate dC_γ of the NaI detector from the isotropic source in the volume element dV is, therefore,

$$dC_\gamma = dV \int_{V_N} \frac{\Gamma S_o(t)}{4\pi R^2} e^{-\mu_1 m(\ell,\beta)} e^{-\mu_2 T(\ell,\beta)} B \, e^{-\mu_3 r} \mu_3 dV_N \quad (A\text{-}2)$$

where $B = 1 + \mu_1^{sc} m(\rho,\beta)$ is the buildup factor, where μ_1^{sc} is the linear total attenuation coefficient for 0.7-Mev gammas in water less the Compton energy absorption coefficient.

Because of symmetry of shape and location of the crystal,

$$dV = 2\pi \ell^2 \sin\theta \, d\ell \, d\theta$$

$$dV_N = 2\pi R^2 \sin\beta \, dR \, d\beta$$

$$RdR = rdr$$

Then,

$$dC_\gamma = dV \frac{\Gamma S_o(t)}{2} \int_{\beta=o}^{\beta_m} \int_{r=o}^{r(\ell,\beta)} \sin\beta \; B e^{-\mu_1 m(\ell,\beta)} e^{-\mu_2 T(\ell,\beta)} \mu_3 e^{-\mu_3 r} dr \, d\beta \qquad (A-3)$$

Because of the complexity of Equation A-3, the assumption is made that the total activity $\Gamma A(t) = \Gamma \int S_o(t) dV$ is located at some distance $\bar{\rho}$. Substituting A(t) and performing the first integration of Equation A-3 gives the total count rate of

$$C_\gamma = \frac{\Gamma A(t)}{2} \int_{\beta=o}^{\beta_m} \sin\beta \; B e^{-\mu_1 m(\beta)} e^{-\mu_2 T(\beta)} \left[1 - e^{-\mu_3 r(\beta)} \right] d\beta \qquad (A-4)$$

with

$$\beta_m = \sin^{-1}\left(\frac{r_o}{a}\right) \quad , \quad \text{where} \quad a = \bar{\rho} + T + r_o$$

$$r(\beta) = 2r_o \left[1 - \left(\frac{a}{r_o}\right)^2 \sin^2\beta \right]^{1/2}$$

$$m(\beta) = a\cos\beta - \sqrt{a^2\cos^2\beta - (a^2 - b^2)} \quad , \quad \text{where } b = r_o + T$$

$$T(\beta) = \sqrt{a^2\cos^2\beta - (a^2 - b^2)} - \sqrt{a^2\cos^2\beta - (a^2 - r_o^2)}$$

The quantity $\bar{\rho}$ is defined by

$$\bar{\rho} = \frac{\int_{\theta=o}^{\theta_m} \int_{\rho=o}^{\rho(\theta)} \frac{1}{4\pi\rho^2} \rho\, e^{-\mu_3 \rho}\, dV}{\int_{\theta=o}^{\theta_m} \int_{\rho=o}^{\rho(\theta)} \frac{1}{4\pi\rho^2} e^{-\mu_3 \rho}\, dV} \qquad \text{(A-5)}$$

Since $dV = 2\pi \ell^2 \sin\theta\, d\ell\, d\theta$, and substituting $\rho\, d\rho$ for $\ell\, d\ell$, performing the first integration of Equation A-5 gives

$$\bar{\rho} = \frac{1}{\mu_3} \frac{\int_{\theta=o}^{\theta_m} \sin\theta \left\{ 1 - \left[1+\mu_3 \rho(\theta)\right] e^{-\mu_3 \rho(\theta)} \right\} d\theta}{\int_{\theta=o}^{\theta_m} \sin\theta \left[1 - e^{-\mu_3 \rho(\theta)} \right] d\theta} \qquad \text{(A-6)}$$

with

$\rho(\theta) = M \sec\theta - b$ for $0^o \le \theta \le \theta_1$

$\rho(\theta) = R_2 \csc\theta - b$ for $\theta_1 < \theta \le \pi/2$

$\rho(\theta) = (R_2 - R_1) \csc(\pi - \theta)$ for $\pi/2 < \theta \le \theta_2$

$\rho(\theta) = (L-M) \sec\theta - R_1 \csc\theta$ for $\theta_2 < \theta \le \theta_m$

where $\theta_1 = \tan^{-1}\left(\frac{R_2}{M}\right)$

$\theta_2 = \pi - \tan^{-1}\left(\frac{R_2}{L-M}\right)$

$\theta_m = \pi - \tan^{-1}\left(\frac{R_1}{L-M}\right)$

By graphical integration of Equation A-6, and using the dimensions previously described, it is determined that

$$\bar{\rho} = 4.9 \text{ cm}$$

The relationship of $\bar{\rho}$ and the NaI crystal is better represented by Figure A-3. For 0.7-Mev gamma rays, μ_1, μ_2, and $\mu_3 = 0.083 \text{ cm}^{-1}$, 0.195 cm^{-1}, and 0.27 cm^{-1}, respectively. μ_1^{sc} is 0.05 cm^{-1}.

Using dimensions and values previously given, and with reference to Figure A-3, Equation A-4 is graphically integrated. As a result,

$$C_\gamma = (2.4 \times 10^{-2})A(t)\Gamma$$

For $\Gamma = 1$ gamma/disintegration and $A(t) = 2.5 \times 10^6$ disintegrations/min, then

$$C_\gamma = 6.0 \times 10^4 \text{ counts/min}$$

The experimentally determined count rate for the stated conditions was 5.8×10^4 counts/min, which is in good agreement.

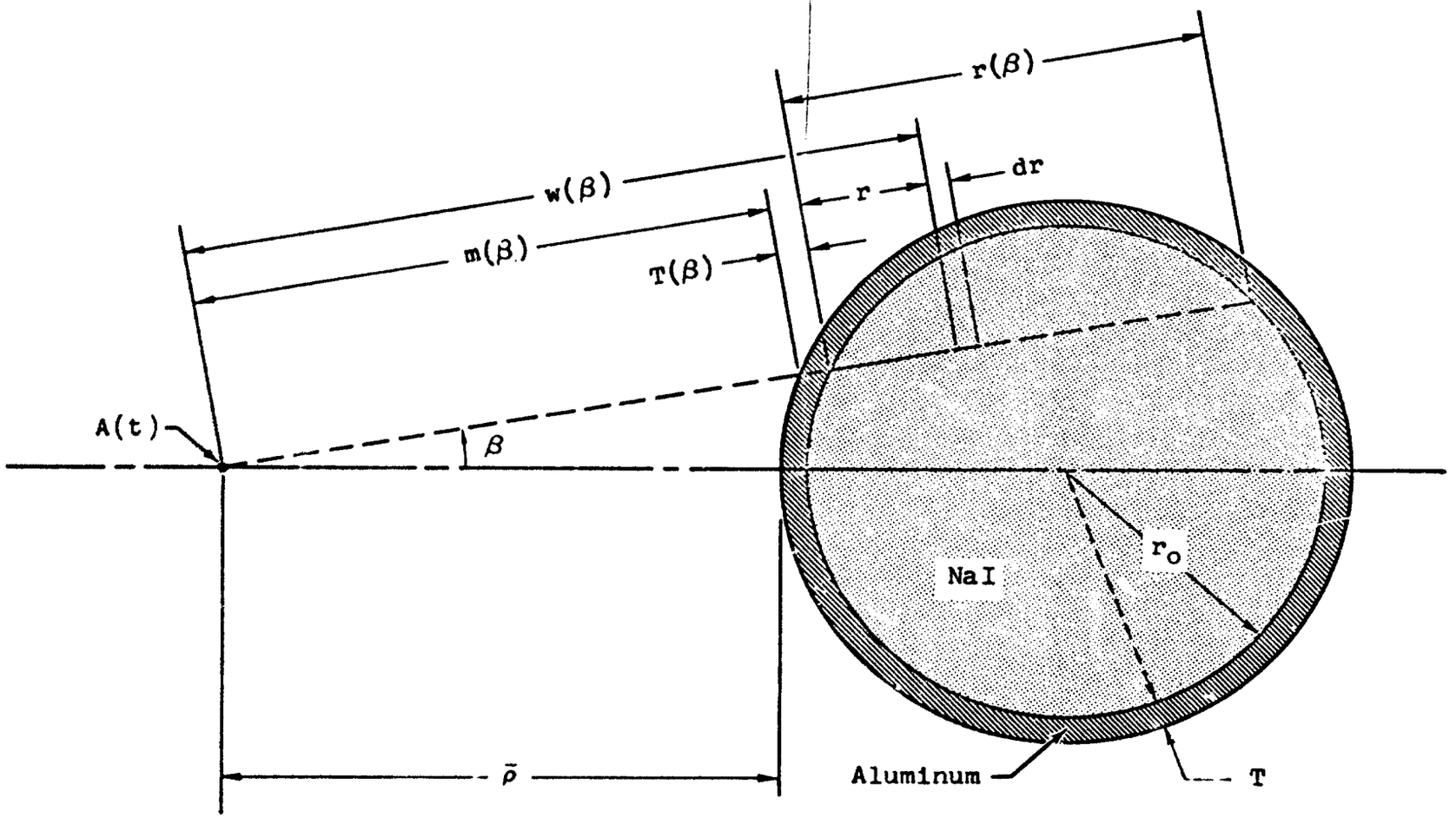
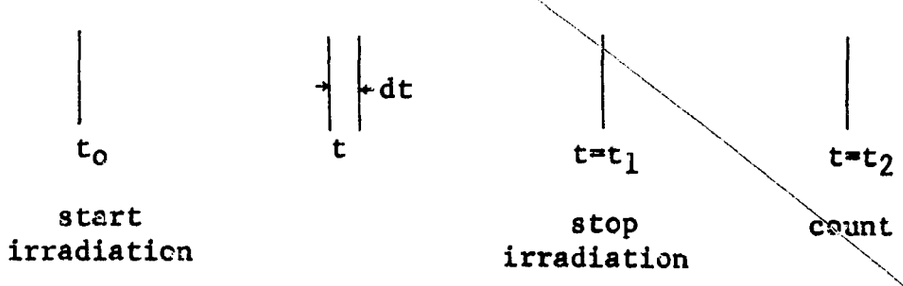


Figure A-3 Simplified Geometry

APPENDIX B

DERIVATION OF EQUATION 4

Consider the following time designation:



The fission rate F is 3.24×10^4 fission/sec.

The number of fissions in time increment dt is F dt .

The number of iodine atoms formed of the $i^{\underline{th}}$ isotope in time increment dt is γ_i F dt .

Each $i^{\underline{th}}$ isotope has a probability P_i of emitting a gamma in the range of interest (Ref. 7).

Of the atoms of the $i^{\underline{th}}$ isotope of iodine formed in time increment dt, at time t_1 , only the fraction $e^{-\lambda_i(t_2-t_1)}$ remains at the count time t_2 .

The total count rate is formed by an integration from t=o to $t=t_1$. That is, if $\gamma_i F dt$ is the number of atoms of the $i^{\underline{th}}$ isotope formed at time t in increment dt, then

$$e^{-\lambda_i(t_2-t)}$$

represents atoms available at count time from dt,

and $$\int_o^{t_1} \gamma_i \, F \quad e^{-\lambda_i(t_2-t)} \, dt$$

represents atoms available at t_2 ,

and $$\lambda_i \int_o^{t_1} \gamma_i \quad F \quad e^{-\lambda_i(t_2-t)} \, dt$$

represents count rate ,

and $$A(T)_i = P_i \, \gamma_i F \int_o^{t_i} e^{-\lambda_i(t_2-t)} \, \lambda_i dt$$

represents count rate in the region of interest, i.e., 820 ± 50 kev.

The integration is

$$A(T)_i = P_i \, \gamma_i F \left[e^{-\lambda_i(t_2-t)} \right]_o^{t_1}$$

$$= P_i \, \gamma_i F \left[e^{-\lambda_i(t_2-t_1)} - e^{-\lambda_i t_2} \right]$$

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13. ABSTRACT

An investigation was conducted on improved methods of fission-product monitoring of the test reactors at the Nuclear Aerospace Research Facility (NARF), Fort Worth Division of General Dynamics. A literature review and the results of an experiment indicated the cascaded resin-bed system as the best method. In this system, a filter removes particulate matter, a cation bed removes most of the activity attributed to corrosion products, an anion b ed removes fission products present in anionic form. In the case of a fission break, the principal activity collected in the anion bed results from the isotopes I-132 and I-134. A single-channel rate meter is employed to monitor a specific range of energies (820 ± 50 kev) corresponding to the most prominent gamma rays emitted by I-132 and I-134. Fission products can be identified by gross-gamma monitoring and their decay rates compared with the Way-Wigner equation. The delay in making this comparison is not in keeping with the fast response desired at NARF, and the system will not be sensitive to small pinhole leaks. A cladding failure of 1-mm^2 area for the Ground Test Reactor is postulated. Analytical results indicate that the activity attributed to the I-132 released from this area would be about equivalent to the background reading taken for a 162-Mw-hr run.

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