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REPORT

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RADIOCHEMISTRY DURING START-UP AND EARLY OPERATION OF THE NUCLEAR SHIP SAVANNAH

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RADIOCHEMISTRY DURING START-UP AND EARLY OPERATION OF THE NUCLEAR SHIP SAVANNAH

by

L. Battist, W. S. Winnowski,
D. K. Dieterly and R. C. Koch

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NUCLEAR SCIENCE AND ENGINEERING CORPORATION
P.O. Box 10901   Pittsburgh 36, Pa.

TABLE OF CONTENTS

ACKNOWLEDGMENT

I. INTRODUCTION

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An experimental program has been completed during the period of the initial testing, start-up, sea trials, and acceptance tests of the N. S. Savannah in which the radioactives in the primary coolant system of the reactor plant were characterized quantitatively. This program was performed for the New York Shipbuilding Corporation under Subcontract 529-2082 who had overall responsibility for the building and testing of the reactor plant and the vessel for the U. S. Maritime Administration, U. S. Department of Commerce, in cooperation with the U. S. Atomic Energy Commission.

The principal objectives of this program were the characterization of the important radioactive species in the primary coolant of the reactor plant and the correlation of their concentrations with important reactor and primary system operating parameters and with radiological safety procedures. In particular, the experimental program was designed to determine, or provide data for the evaluation of, the reliability or performance of specific components or systems of the shipboard reactor plant. Data were required to evaluate the integrity of fuel element cladding, to estimate the uranium contamination of heat transfer surfaces in the core, corrosion in primary system surfaces, and to monitor the performance of the demineralizer, off-gas, and water-purification systems. Additional information was needed to determine base levels for the radioactivity content of the primary coolant to be used in evaluating data acquired during subsequent reactor operations; to detect any leakage of radioactivity from the primary to secondary coolant systems; and to assure that radioactivity concentrations in liquid wastes would not exceed the maximum permissible concentrations established by Federal and state agencies.

Substantial information for primary coolant system technology of pressurized water reactors was available prior to the inception of this program. This program was formulated[1,2] on the basis of the existing technology with introduction of those modifications normally required for a shipboard reactor and by the specifications set forth for the N. S. Savannah[3].

Three general types of radionuclides comprise the radioactivity content of the primary coolant: (1) fission products, (2) neutron-activated corrosion products, and (3) activation products formed in coolant constituents and impurities. Fission products are introduced into the coolant either as the result of defective fuel elements, of direct fission-product recoil from traces of uranium on structural or fuel cladding surfaces, or of neutron-induced fission in trace quantities of fissile elements in the primary coolant. The activated corrosion products result from the interaction of neutrons with minute quantities of structural materials which are transported through the core by the primary coolant. Other activation products are formed as the result of the interaction of the neutrons or of secondary nuclear particles (e. g., protons) with traces of impurities in the primary coolant or with constituents of the coolant itself. Typical radionuclides of each type which are expected to be present in significant concentrations in the primary coolant of a pressurized water reactor are shown in Table I. These sources of primary coolant contamination are discussed below.

A. Fission Products

Data for the concentrations of selected fission products may be utilized to determine the amount of uranium contamination on core surfaces, to detect any defects in fuel cladding, and to estimate the severity of any failure of a fuel element cladding. Under normal operating conditions, uranium in the primary coolant is at very low concentrations, and thus is not expected to be a significant source of fission products in the coolant. If the uranium concentration is known and small, the fission product concentrations arising from this source can be computed with an accuracy sufficient for use as correction factors in the application of the fission product concentration data to evaluate other plant parameters.

The concentrations in the primary coolant of short-lived fission products arising from recoils from uranium contamination on core surfaces are generally proportional to the mean neutron flux in the core. Therefore, at steady-state reactor operations (i. e., the condition established when the rate of recoil of a fission product into the coolant equals its rate of decay plus the rate of removal by the demineralizer), the concentration of a given species is expected to be constant. It is also insensitive

TABLE 1

TYPICAL RADIONUCLIDES PRESENT IN PRESSURIZED WATER REACTOR COOLANT

Coolant-Activation Products		Fission Products		Activated Corrosion Products	
N-16	7.35 seconds	I-134	52.5 minutes	Mn-56	2.57 hours
N-13	10.0 minutes	I-135	6.75 hours	Ni-65	2.56 hours
Ar-41	110 minutes	I-133	20.8 hours	Cu-64	12.8 hours
F-18	112 minutes	I-131	8.05 days	W-187	24.0 hours
Na-24	15.0 hours	Cs-139	9.5 minutes	Cr-51	27.8 days
		Cs-138	32.2 minutes	Fe-59	45.1 days
		Cs-137	30 years	Zr-95*	65 days
		Ba-139	83 minutes	Co-58	71.3 days
		Ba-140	12.8 days	Co-60	5.24 years
		Sr-91	9.67 hours	Mn-54	308 days
		Sr-92	2.7 hours		
		Sr-89	50.5 days		
		Sr-90	27.7 years		
		Zr-95	65 days		
		Mo-99	66 hours		
		Xe-133	5.27 days		
		Xe-135	9.2 hours		
		Xe-135m	15.3 minutes		
		Kr-85	10.3 years		

* from follower rod assembly

to operating time and to reactor power transients once the steady-state condition has been achieved. This condition is established after operation of the reactor at a constant power with constant coolant and let-down flow rates for a period of approximately four half-lives of the fission product. Furthermore, the concentrations of short-lived fission products arising from this source are expected to be proportional to their fission yields. The method of calculating the uranium contamination of the core surface is discussed in Appendix I.

The concentrations in the primary coolant of fission products resulting from leakage through a defective fuel element cladding generally exhibit different patterns. For example, the release rate of a given radionuclide may be a function of its volatility or solubility in the coolant, of the volatility or solubility of a radioactive precursor, of the temperature or burnup history of the exposed fuel, or of the size of the breach in the cladding. The ratios of these concentrations, therefore, generally are insensitive to the fission yields. Although the concentrations will normally vary with reactor power, pronounced changes in concentration frequently occur as a result of power transients or following start-up or shutdown of the reactor.

Fission products which are volatile at fuel element temperatures are known to be released preferentially from the fuel. The noble gas fission products, or their short-lived decay products (e. g., isotopes of rubidium, strontium, cesium, and barium), and isotopes of volatile elements such as iodine and bromine may be employed as sensitive indicators of a cladding defect or failure. The behavior of the concentrations of two or more of such fission products as a function of time and reactor operating parameters can thus be used to detect rapidly any significant cladding defect.

If the concentration of these fission products in the coolant resulting from uranium contamination of core surfaces has been established, these values may be used as a base line for normal reactor operations. Significant deviations from the base line concentrations then indicate that a fuel element cladding may have become defective. One phase of the experimental program was performed to determine the "normal" fission product concentrations and the uranium contamination of core surfaces.

The selection of the optimum fission products for routine surveillance of the primary coolant was based on several considerations. It is desirable that they be formed in high yield in the fission process, that they have short half-lives, and that the requisite analyses can be performed rapidly and accurately. It is important that they be preferentially released from the fuel to the coolant, and that they be soluble in the coolant under plant operating conditions. Many fission-product elements, such as tellurium, cerium, and zirconium, tend to deposit, in varying degrees, on particulate matter in the coolant or on primary system surfaces. Such nuclides should thus not be selected as sensitive indicators of cladding failures.

The radioisotopes of iodine satisfy the principal criteria for cladding failure indicators. Therefore, a special rapid method of analysis for iodine radioactivity was selected for routine surveillance of fission product concentrations in the primary coolant.

Some radioactive fission products are important also from a radiological safety viewpoint. For example, Sr^{90}, by virtue of its long nuclear and biological half-lives, is of serious concern if it is introduced into the biosphere. Controls must be established, therefore, to prevent release to the environment of any plant effluents which may contain a quantity of a radioactive species leading to environmental contamination levels in excess of its maximum permissible concentration. For a reactor plant, information is also required to evaluate the radiological hazard which might result from leakage or other inadvertent releases of contained fluids. Therefore, data for the normal concentrations of the longer-lived fission products in reactor coolants and in liquid wastes prior to discharge are required.

B. Activated Corrosion Products

At the normal operating temperature range of pressurized water reactors, corrosion of the structural materials of the primary system proceeds at a slow but finite rate. Minute quantities of these materials enter the coolant and are present in true solution, or in colloidal or particulate forms. The particulate fraction is commonly referred to as crud. These corrosion products are circulated through the core by the coolant. As a result, neutron-activated corrosion products are formed. These radionuclides also may be present in solution or as colloids, or they may be associated with the crud fraction.

The crud fraction tends to deposit on primary system surfaces, where the contained longer-lived activated corrosion products may constitute a radiological hazard in the event that access is required for routine or emergency maintenance operations.

The distribution of the crud between a liquid suspension and a deposited form depends on the size of the particulates, the specific materials from which the system is fabricated, the temperature, and the flow rate and pH of the coolant. The latter parameter is also an important factor in determining the distribution of the activated corrosion products (and of some fission products) between the liquid and solid phases in the primary coolant. For example, in the typical range of pH values of pressurized water reactor coolants (pH 7 - 11), the major fractions of the activated corrosion products generally are associated with the crud fraction. Therefore, it is essential to determine the concentrations of these radionuclides in both phases of the coolant to obtain the desired data for corrosion and for potential contamination of system surfaces.

After sustained operation of the reactor, and in the absence of a defective fuel element cladding, the activated corrosion products usually constitute the predominant long-lived radioactivities in the primary coolant. It is important that the demineralizer system be effective for these species to minimize the potential contamination of system surfaces and to assure that their concentrations in liquid effluents are consistent with radiological safety requirements. Periodic measurements of activated corrosion products in the influent and effluent of the demineralizer provide tests of its performance in removing these important radioactive species.

C. Coolant-Activation Products

The third type of radioactive species present in the primary coolant results from interaction of neutrons or gamma-rays or of secondary particles (i. e., protons, alpha particles) with impurities in the coolant or with coolant constituents. For example, N^{16} may be produced from the reaction $O^{16}(n, p)N^{16}$; N^{13} is believed formed by the reaction $O^{16}(p, \alpha)N^{13}$ [4]; F^{18} by the reaction $O^{18}(p, n)F^{18}$; Ar^{41} by the reaction $Ar^{40}(n, \gamma)Ar^{41}$; and Na^{24} by the reaction $Na^{23}(n, \gamma)Na^{24}$. Radionuclides such as N^{16} and F^{18} are thus intrinsic to aqueous primary coolants or moderators.

Dissolved air leads to production of Ar^{41}, and Na^{24} is formed from sodium impurities in the primary coolant. Therefore, these latter two species can be used as indicators, respectively, of excessive air or cation impurities in the primary system. Ar^{41} is particularly useful in detecting air leakage into the sytem or inadequate removal of air during purification of make-up water. Since oxygen has a strongly deleterious effect on corrosion rates in water-cooled reactors, Ar^{41} analyses serve an especially useful purpose in coolant quality control.

Nitrogen-16 emits very energetic gamma radiations and is a prime consideration in reactor shielding. Excessive concentrations of Na^{24} may also introduce stringent radiation-shielding requirements. Concentrations of these species are thus of interest from a radiological safety viewpoint. Early indication of gross increases in their concentrations can permit corrective action to be taken to reduce their concentrations before they represent radiological safety or engineering problems.

The radiochemistry studies performed during the start-up and initial operations of the reactor plant aboard the N. S. Savannah provided the data to assist in evaluating the various parameters cited above. The experimental program and experimental results are presented in the next two sections, respectively, and a discussion of the results is given in the final section.

II. EXPERIMENTAL PROGRAM

II. EXPERIMENTAL PROGRAM

A. General Methods

The experimental program was designed to meet two objectives: (1) daily surveillance of the radioactivity content of the primary system of the reactor plant, and (2) detailed characterization of the radioactivities at various stages of the plant start-up and test operations. Routine surveillance operations involved daily measurements designed to indicate rapidly changes in the gross quantity of radioactivity in the primary system and to monitor the coolant for critical constituents. Three types of measurements were performed on primary coolant samples in these operations: (a) analyses of "gross 15-minute degassed activities", (b) gross one-hour iodine analyses, and (c) gamma-spectrum analyses of one-liter samples of the unfiltered primary coolant.

The gross 15-minute degassed activity was determined by degassing a 2-ml aliquot of primary coolant and gamma counting it exactly 15 minutes after sampling. All gamma radiations having energies greater than 30 Kev were measured. This measurement served as a rapid indicator of the gross radioactivity in the primary coolant.

The gross one-hour iodine activity was determined in duplicate one-liter samples of filtered coolant by performing chemical separation of iodine and determining the gamma activity in the iodine fraction exactly one hour after sampling. This is a rapid method of estimating fission product concentrations in the primary system and of detecting fuel cladding failure.

Periodically, integral decay curves were obtained for both types of samples. These curves represent the gross decay of all radionuclides in the samples which emit gamma radiations having energies greater than 30 Kev. The observation of the gross gamma spectrum of the coolant served as an indicator of changes in the concentrations of its principal radioactive constituents. This series of analyses was deemed to be the minimum requirement for surveillance of the overall performance and reliability of the primary system.

Periodically, samples of secondary and intermediate system fluids were

analyzed to determine that no radioactivity had leaked into these systems. The liquid waste tanks were also assayed to assure that discharged liquids contained no more than the maximum permissible concentration of radioactivity.

During periods of sustained reactor operations at relatively constant power, detailed characterizations of the radioactive species in the primary coolant were performed. These characterizations involved the quantitative determinations of individual radionuclides in the coolant and of their distributions between the aqueous, solid, and gas phases, if possible. The distribution of these radionuclides between the aqueous phase (solubles) and solid phase (corrosion product particulates, or crud) was determined by measurements on one-liter samples of filtered and degassed coolant and on particulate samples removed from the coolant during the filtering operation. Unfortunately, direct measurement of gas-phase radioactivity could not be performed because the shipboard gas-sampling system was inoperative for the duration of this program. Estimates of this activity were attempted, however, from data obtained in other samples. In addition, the efficiency of the demineralizer was determined periodically by measurement of specific radionuclides in samples of influent and effluent coolant.

Radioactivity balances were also performed during periods of constant reactor power. These measurements involved the search for all significant radioactive constituents in a sample of primary coolant and the comparison of the summation of the radioactivity in the chemically separated fractions with the gross gamma radioactivity in degassed samples of coolant. These measurements assured that all gamma-emitting radionuclides present in significant quantities of the primary coolant were identified and measured.

All samples were removed from the reactor system under the supervision of an NSEC representative and of the shift health physicist. The samples of primary coolant were taken at the demineralizer influent at a constant flow rate of one liter per minute. The samples were immediately filtered and/or degassed, as required, in preparation for analytical measurements.

The analytical procedures used in these studies were based on proven methods[5,6,7,8]. These procedures were modified, when necessary, to meet the

specific requirements of this program.

The basic experimental program may be conveniently divided into two sections: (1) operations at Camden, corresponding to Phases III and IV-A for the construction and testing of the vessel[1], and (2) operations at Yorktown, corresponding to Phases IV-B and V.[2] Descriptions of the studies performed are now presented.

B. Operations at Camden

1. Pre-Criticality Activities (Phase III)

At the inception of this program, the need for suitable laboratory space and facilities aboard ship was recognized. Such facilities included a laboratory for radiochemistry processing and a separate area for the nucleonics instruments used for measurement of the radioactivities in processed samples. The lack of such facilities, either aboard ship or in the shipyard, necessitated construction of suitable laboratory space. NSEC personnel made recommendations for the required laboratory facilities and equipment.

Recommendations for the radiochemistry laboratory were based on the minimum space requirements of the four-man staff assigned to perform the on-site program and on the nucleonics equipment specified to conduct the experimental studies. Two hundred square feet of laboratory space, containing two fume hoods, approximately twenty linear feet of laboratory bench space, normal laboratory utilities, and radioactive waste disposal facilities, was judged to be a minimum requirement to perform properly the experimental program. An additional space of at least 100 square feet was recommended for installation of the nucleonics instruments and for desk space for data processing. After the decision to construct both the laboratory and instrument room in trailer bodies, NSEC personnel prepared design criteria consistent with the above recommendations, selected the required equipment, assisted in the construction and installation operations, and procured the necessary laboratory ware, supplies, and reagents.

The following NSEC-owned radioactivity measuring equipment was provided for this program:

(1) a fully transistorized 256-channel, pulse-height analyzer, with a

3-in. x 3-in. NaI(Tl) crystal, analogue data display, and digital read-out;

    (2) a single-channel, pulse-height analyzer-scaler assembly, with a 1-3/4-in. x 2-in. NaI(Tl) well-type crystal; and

    (3) a scaler, power supply, and argon-filled, halogen-quenched geiger tube for beta counting.

This equipment was installed in the instrument trailer and was calibrated for its intended use.

During this portion of the program, a set of samples was obtained and analyzed to determine the extent, if any, of radioactive contamination of the reactor system prior to initial criticality. Such contamination might have resulted from fresh fallout from the 1961 U.S.S.R. nuclear weapons test series, or from radioactitives created in the reactor core during pre-installation criticality experiments and released to the fresh coolant through gross defect in the fuel cladding.

Samples were withdrawn from the primary, secondary, intermediate, and other auxiliary systems as detailed in Table III (p. 23). These samples were analyzed in the NSEC laboratories for gross alpha, gross beta, and gross gamma radio-activities, for K^{40}, Sr^{90}, and Cs^{137}, using ultra-sensitive radioactivity measurement methods, and for uranium (fluorimetrically).

2. Early Power Operations (Phase IV-A)

Immediately after initial criticality of the reactor was achieved, the experimental radiochemistry program was initiated. Routine surveillance of the primary system was established and continued during all periods in which the reactor was critical. The routine analyses cited previously were performed throughout this phase.

During this phase, the reactor operated for a sustained period at 9.7% of full power.* A detailed characterization of the radioactivities in the primary coolant was performed at this power level. This experiment involved measurement of N^{13}, F^{18}, Na^{24}, Mn^{56}, $Sr^{91, 92}$, $I^{133, 134, 135}$, Cs^{138}, and Ba^{139} in a filtered and de-gassed sample of primary coolant. The separated crud fraction was assayed by

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* For purposes of these studies, full power was taken to be 69 Mw(th).

gamma spectrometry and gross decay measurements.

C. Operations at Yorktown (Phases IV-B and V)

The N. S. Savannah was removed to the U. S. Coast Guard Reserve Training Center at Yorktown, Virginia, for test operations at power levels in excess of 10% of full power. This move necessitated partial dismantling of both the laboratory and instrument trailers. When the move had been effected, the trailers were restored to operating conditions.

The radiochemistry program was resumed when the reactor was again at power. Surveillance of the primary system was maintained, as described previously, at all times when the reactor was operating. Periodic surveillance of intermediate, secondary, and waste disposal systems was performed. Detailed studies of the radio-activities in the primary system were conducted at a series of power levels during dockside operations and during power tests at sea, including the sea trials, and the acceptance tests. These studies, which included radioactivity balance measure-ments, were performed at average power levels corresponding to 18%, 38.9%, 53%, 77.6%, and 88.7% of full reactor power. At power levels below 38.9%, analyses were performed for Ni^{65} and W^{187} in addition to those radionuclides sought in the experiments performed at 9.7% of full power. At higher power levels, analyses were conducted for longer lived species such as Cr^{51}, Mn^{54}, Fe^{59}, $Co^{58,60}$, I^{131}, Ba^{140}, and, on occasion, Sr^{89} and Zr^{95}. Thus data were obtained for important fission products, for activation products resulting from coolant constituents or impurities, and for activated corrosion products.

III. EXPERIMENTAL RESULTS

III. EXPERIMENTAL RESULTS

A. Plant Surveillance

The concentrations of gross gamma radioactivity and of short-lived radioactive fission products in the primary coolant during early reactor operations were expected to be directly proportional to the reactor power level. Therefore, all experimental values for these parameters were normalized to full reactor power to provide a basis for comparison of data obtained in sequential experiments.

The concentration of the normalized gross 15-minute degassed activity was observed to have an average value of $(5.6\pm0.8) \times 10^{-2}$ $\mu c/ml$ at power levels under 10% of full power. This mean value increased with increasing reactor power and became essentially constant at $(1.2\pm0.2) \times 10^{-1}$ $\mu c/ml$ at power levels greater than 18% of full power. This latter value has been taken as the base operational level for the gross 15-minute degassed activity. A typical decay curve of a gross 15-minute degassed sample is shown in Figure 1.

During operation of the reactor at less than 10% of full power, the average normalized value for the concentration of the gross one-hour iodine activity was $(1.2\pm0.2) \times 10^{-4}$ $\mu c/ml$. At higher power levels, the average normalized value was $(2.0\pm0.3) \times 10^{-4}$ $\mu c/ml$. This latter value has been taken as the base operational level for the gross one-hour iodine concentration. A typical decay curve obtained in this manner for a gross one-hour iodine sample is shown in Figure 2.

The daily gross gamma spectrum measurements on samples of unfiltered primary coolant were used to determine rapidly the principal radioactive constituents of the coolant. Figures 3 and 4 show typical spectra obtained at reactor power levels of 9.7% and 88.7% of full power, respectively. Generally, during low power operations, the principal radioactivites in the coolant were N^{13}, F^{18}, Ar^{41}, and Mn^{56}. Sodium-24 became increasingly important with increasing reactor operation, as is seen by comparison of Figures 3 and 4. The 1.37-Mev photopeak due to Na^{24} obscured the 1.28-Mev photopeak of Ar^{41} at the higher power level, even though the Na^{24} contribution to the total gamma radiation immediately after sampling was small relative to that of N^{13}, F^{18}, and Mn^{56}.

FIGURE 1

Typical integral gamma decay curve of gross 15-minute
degassed activity.   Reactor at 53% of full power.
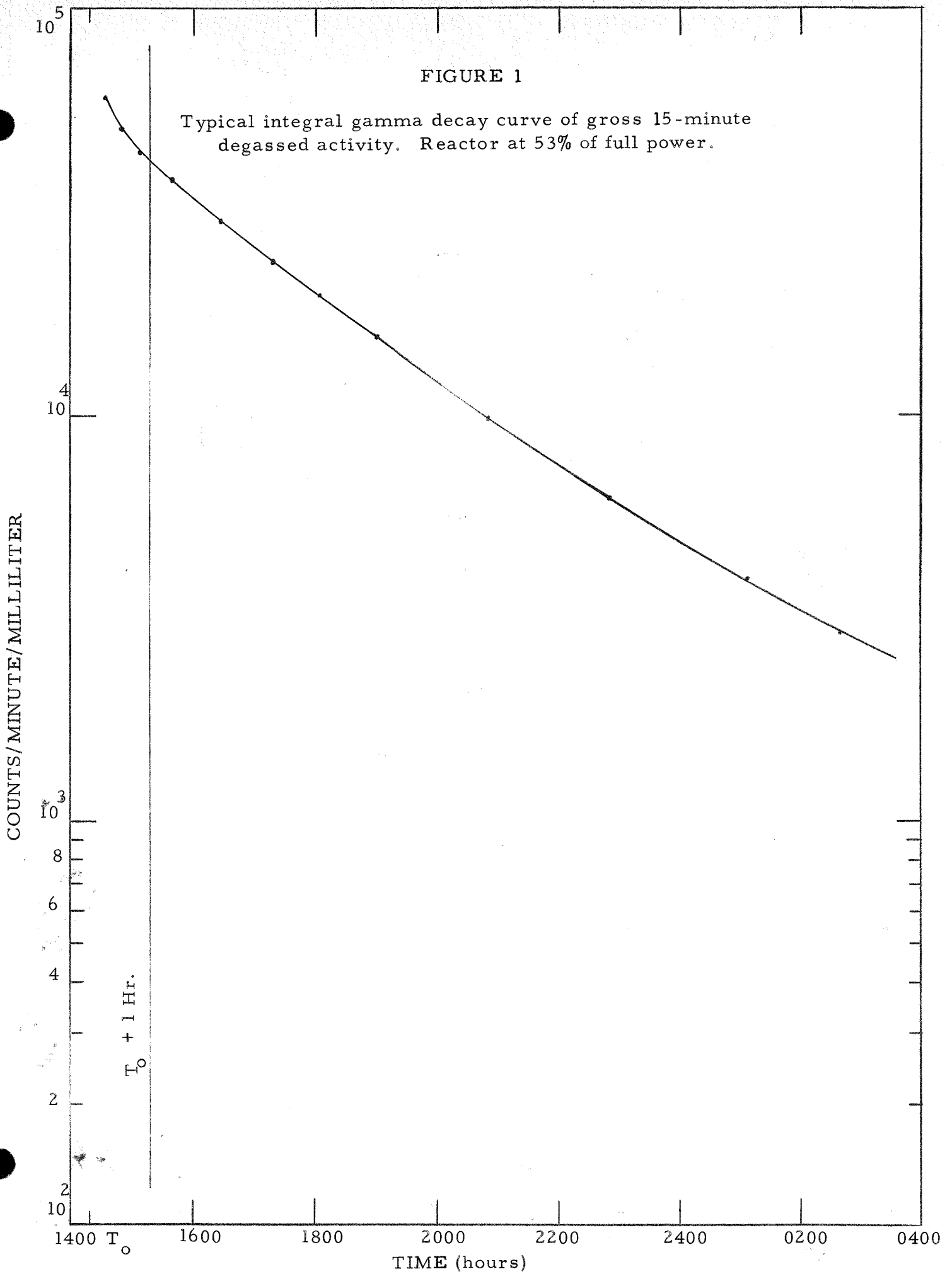
FIGURE 2

Typical integral gamma decay curve of gross
one-hour iodine. Reactor at 53% of full power.

Integral I decay

I^{134}
52.5 minutes

T_O + 1 Hr.

COUNTS/MINUTE/MILLILITER

TIME (hours)
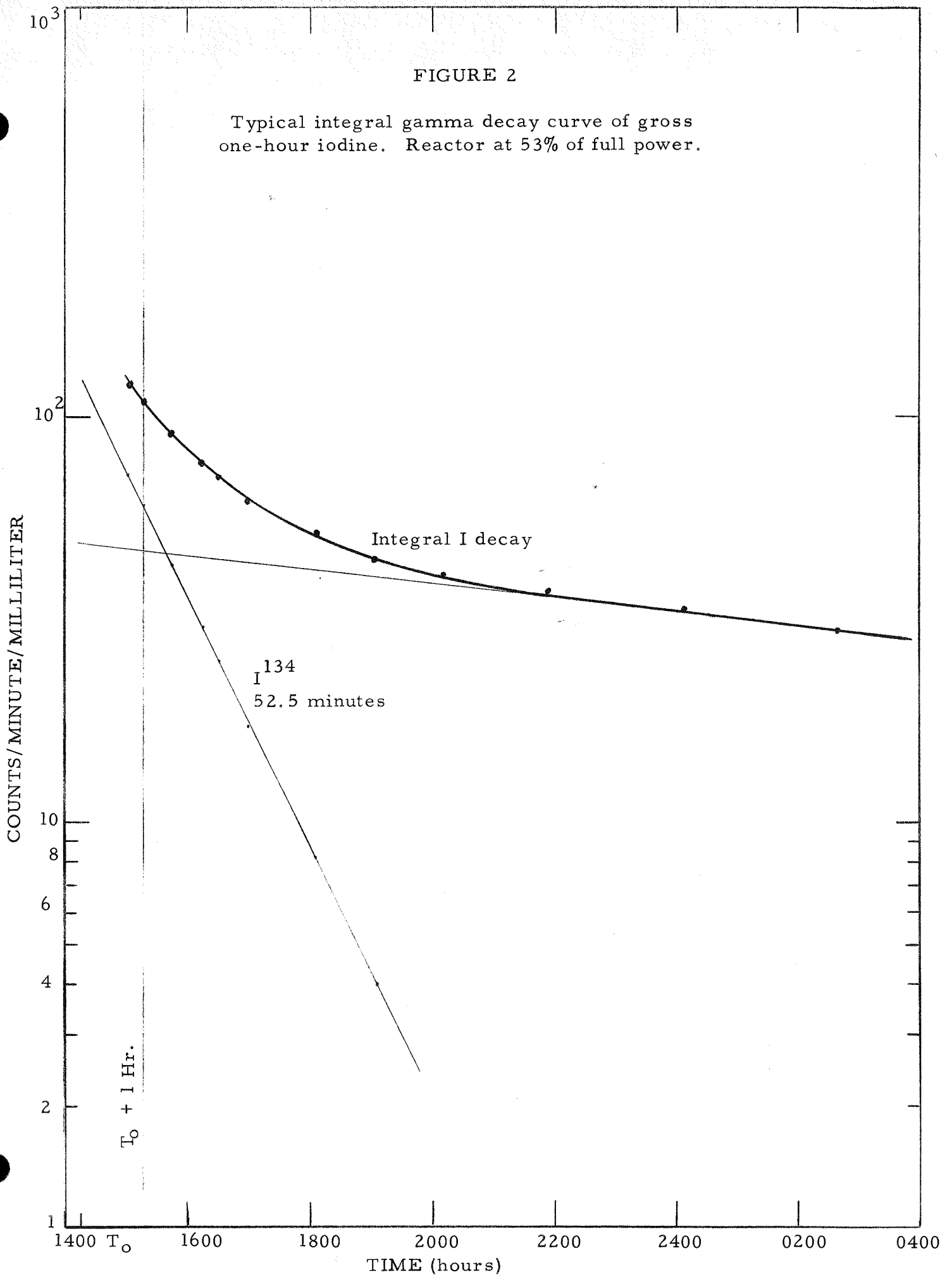
FIGURE 3

Typical gross gamma spectrum of primary coolant. Reactor at 9.7% of full power.

0.51 Mev Positron Annihilation Peak
F^{18}
N^{13}

0.86 Mev Photopeak
Mn^{56}

1.28 Mev Photopeak
Ar^{41}

1.80 Mev Photopeak
Mn^{56}

2.1 Mev Photopeak
Mn^{56}

COUNTS/CHANNEL/2 MIN./LITER

CHANNEL (10 Kev/channel)
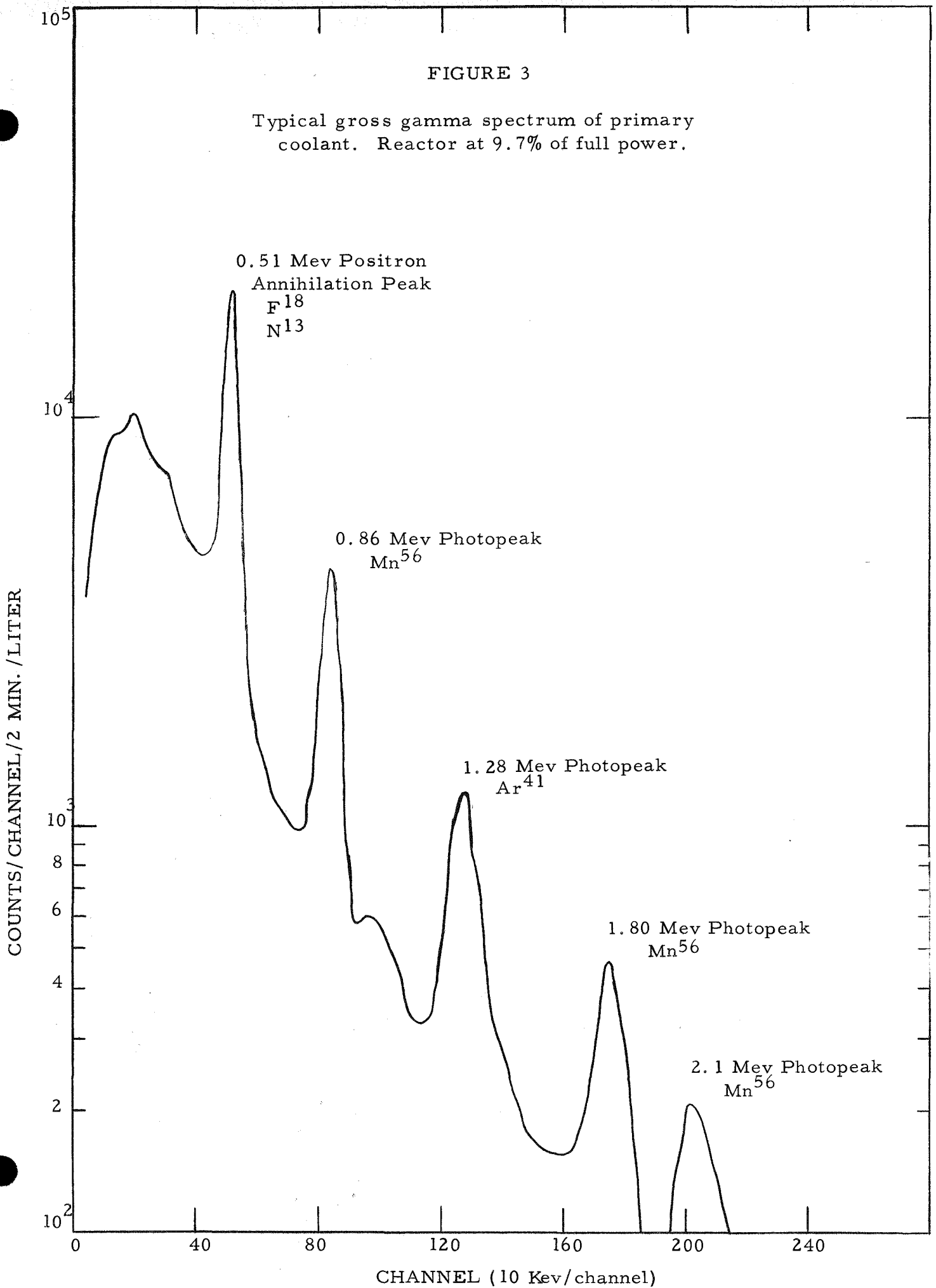
FIGURE 4

Typical gross gamma spectrum of primary coolant. Reactor at 88.7% of full power.
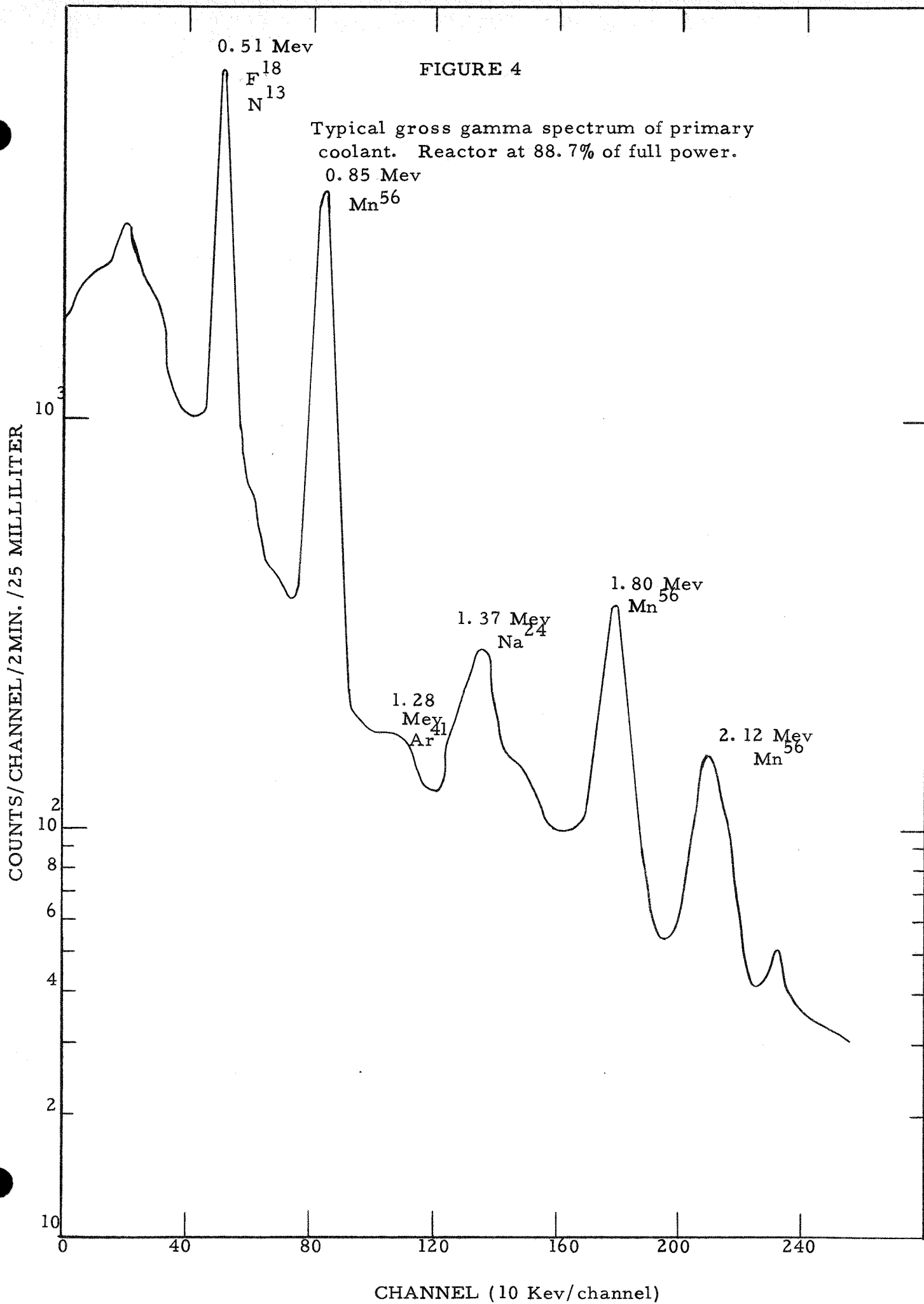
During the course of the surveillance operations, the spectra of selected un-filtered primary coolant samples were observed as a function of time after sampling. Analyses of the decay characteristics of individual photopeaks yielded quantitative values for the principal radionuclides in the coolant. Since 10-minute N^{13} and 112-minute F^{18} decay by positron emission, component analysis of the complex decay of the 0.51-Mev annihilation photopeak was required. The method of analysis is demonstrated in Figure 5. Data for the concentrations of N^{13}, F^{18}, and Ar^{41} as a function of reactor power are shown in Table II.

B. Studies of Specific Nuclides

1. Pre-operational Samples

Results of the analyses of a sample of primary coolant, of fluids from auxiliary systems, and of water sources for the N. S. Savannah are shown in Table III. In general, no significant concentrations were observed for the radioactive species sought.

Table IV contains the concentrations of two long lived fission products, Sr^{90} and Cs^{137}, of gross alpha and beta radioactivities, and of uranium in primary coolant samples taken just prior to initial criticality and as a function of early reactor power levels. Pre-criticality samples were taken at varying coolant temperatures and flow rates. No significant concentrations of the sought radio-activities were observed. The uranium concentration in the primary coolant prior to initial criticality was $(12\pm2) \times 10^{-5}$ mg/l. The uranium concentration exhibited a decreasing trend with continuing reactor operation.

2. Activation Products

Data were obtained at various reactor power levels for the concentrations in filtered and/or degassed primary coolant samples of three radionuclides, N^{13}, F^{18}, and Na^{24}, resulting from activation of coolant constituents or impurities. The values for F^{18} and Na^{24} were obtained by gamma spectrometry measurements on the separated fractions. Typical gamma spectra of the respective separated fractions are shown in Figures 6 and 7. The values reported for N^{13} were derived

FIGURE 5

Component Analysis: 0.51 Mev Photopeak
decay. Reactor at 18% of full power.

112 min.
F^{18}

10 min.
N^{13}

T_o

COUNTS/MINUTE/MILLILITER

TIME (hours)
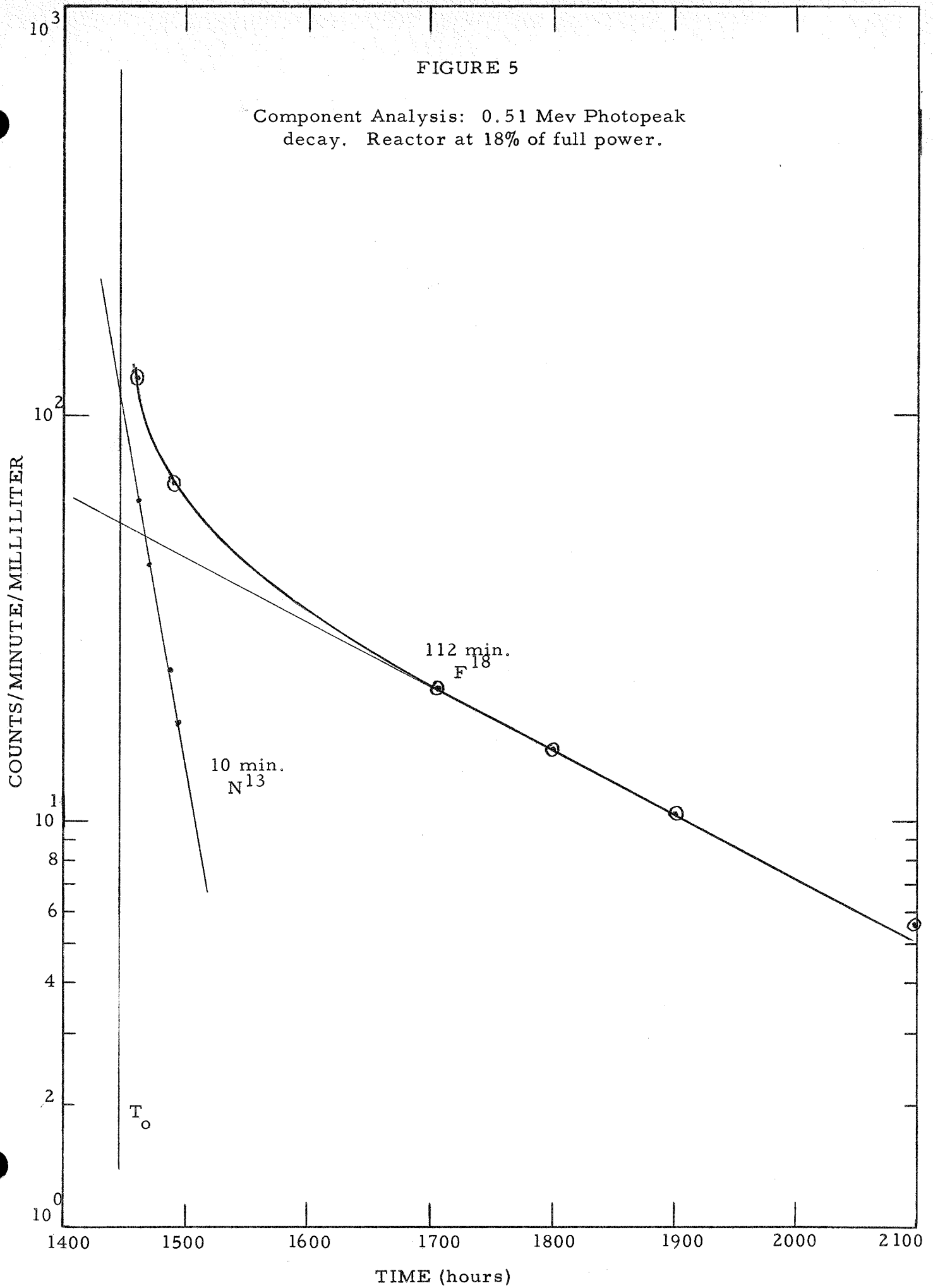
TABLE II

CONCENTRATIONS OF ACTIVATION PRODUCTS IN THE PRIMARY COOLANT [a]

Reactor Power (% of Full Power)	N^{13} Concentrations [b]		F^{18} Concentrations [b]		Ar^{41} Concentrations [b]	
	dpm/ml	μc/ml	dpm/ml	μc/ml	dpm/ml	μc/ml
9.7	8.0×10^3	3.6×10^{-3}	2.2×10^3	9.9×10^{-4}	2.1×10^2	9.5×10^{-5}
18.0	9.8×10^3	4.4×10^{-3}	4.8×10^3	2.1×10^{-3}	4.1×10^2	1.8×10^{-4}
38.9	3.2×10^4	1.4×10^{-2}	1.1×10^4	5.0×10^{-3}	1.9×10^3	8.6×10^{-4}
53.0	5.4×10^4	2.4×10^{-2}	1.6×10^4	7.2×10^{-3}	2.2×10^3	9.9×10^{-4}
77.6	7.1×10^4	3.2×10^{-2}	1.9×10^4	8.5×10^{-3}	2.5×10^3	1.1×10^{-3}
88.7	9.2×10^4	4.1×10^{-2}	2.5×10^4	1.1×10^{-2}	5.0×10^3	2.2×10^{-3}

(a) Concentrations determined by analysis of gross gamma spectra.

(b) Data corrected for decay to time of sampling.

TABLE III

CONCENTRATIONS OF RADIOACTIVITIES IN PRE-OPERATIONAL SAMPLES

Sample No.	Date of Sampling	Gross Alpha[a] (dpm/l)	Gross Beta[a] (dpm/l)	K^{40} (dpm/l)	Gross Gamma (dpm/l)	Sr^{90} [a] (dpm/l)
DFT-1	11-16-61	1.5±0.8	0±2		0	0±0.5
BS-1	11-27-61	2.5±0.9	0±2		0	0±0.5
PV-1	11-26-61	1.0±0.5	12±3	0.04	0	0±0.5
DWM-1	11-16-61	1.9±0.7	0±2		0	0±0.5
BFT-1	11-16-61	1.7±0.9	0±2		0	0±0.5
PSG-1	11-8-61	2.0±0.7	0±2		0	0±0.5
YWW-1	11-15-61	3.4±0.9	20±3	0.05	0	0±0.5
CCW-1	11-15-61	4.5±1.1	20±3	0.05	0	0±0.5
PDW-1	11-15-61	1.2±0.9	1±2		0	0±0.5
GCW-1	11-15-61	2.2±0.9	12±3	0.04	0	0±0.5
DE-1	11-8-61	3.6±0.8	0±2		0	0±0.5
VH-1	11-8-61	0.7±0.8	0±2		0	0±0.6
SSG-1	11-8-61	3.1±0.9	0±2		0	0±0.5

(a) The error designates one standard deviation due to counting statistics.

DFT	=	De-aerator feed-water tank	CCW	=	Camden city water
BS	=	Buffer seal booster pump	PDW	=	Power house distilled water
PV	=	Primary vessel	GCW	=	Gloucester city water
DWM	=	Distilled water makeup tank	DE	=	Demineralizer effluent
BFT	=	Boiler feed-water tank	VH	=	Vent on main discharge header
PSG	=	Port steam generator	SSG	=	Starboard steam generator
YWW	=	Yard well water			

TABLE IV

ANALYTICAL DATA FOR PRIMARY COOLANT SAMPLES
DURING EARLY REACTOR OPERATIONS[a]

Sample No.	Date of[c] Sampling	Gross Alpha[b] (dpm/l)	Gross Beta[b] (dpm/l)	Sr^{90}[b] (dpm/l)	Cs^{137}[b] (dpm/l)	Uranium (mg/l)
PV-2	12-20-61	0±0.8	0±8	0±0.5	0±0.6	0.00012
PV-3	12-20-61	0±0.8	0±8	0±0.7	0.7±0.7	0.00012
PV-4	12-21-61	0±0.8	0±8	0±0.6	1±1	0.00010
PV-5	12-21-61	0±0.8	0±8	0±0.6	0±0.9	0.00018
PV-11	12-28-61	0±0.8	0±8	0±0.9	1±1	0.00008
PV-13	1-3-62	0±0.8	0±8	0±0.8	1±1	0.00006
PV-14	1-10-62	0±0.8	0±8	0±1.2	1±1	0.00008
PV-15	1-11-62	0±0.8	0±8	0±0.7	0.5±1	0.00004

(a) Samples taken at varying conditions of coolant temperature and flow rate and of reactor power.

(b) The error designates one standard deviation due to counting statistics.

(c) Initial criticality was achieved at 1945 hrs, 12-21-61.
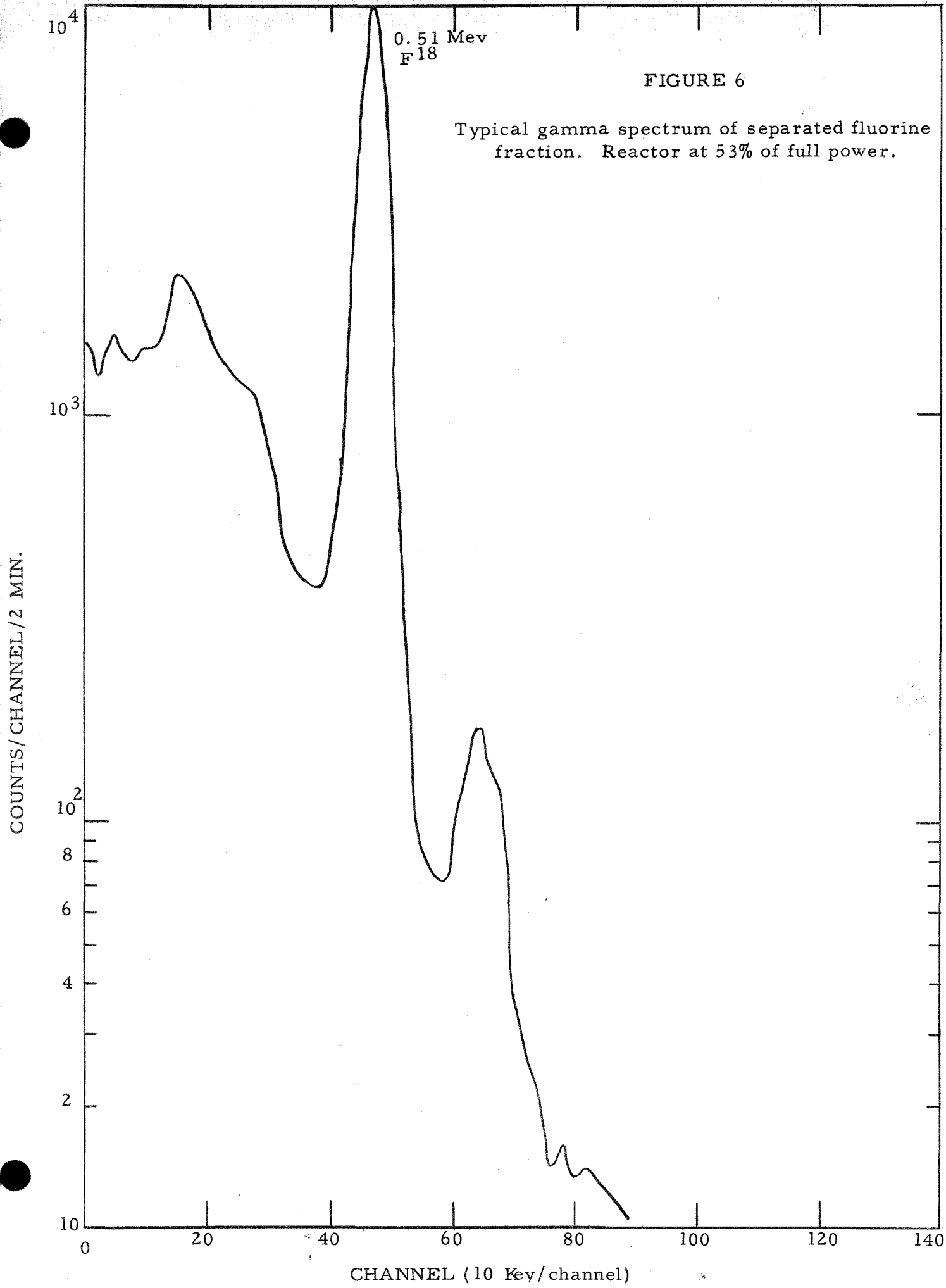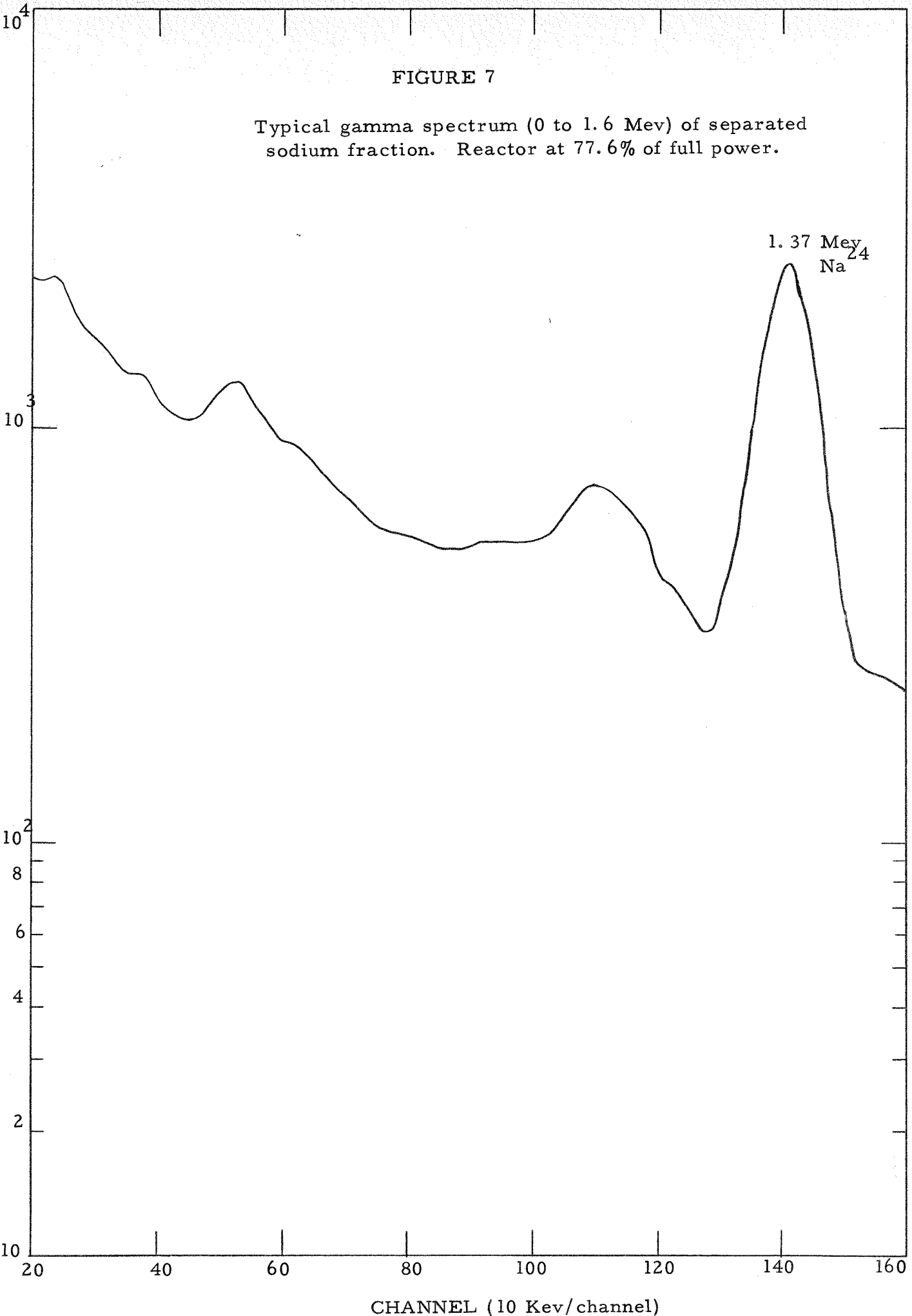
FIGURE 6

Typical gamma spectrum of separated fluorine fraction. Reactor at 53% of full power.

0.51 Mev
F^{18}

FIGURE 7

Typical gamma spectrum (0 to 1.6 Mev) of separated sodium fraction. Reactor at 77.6% of full power.

- 24 -

from component analyses of the gross decay curves obtained from aliquots of degassed coolant samples and thus represent only those fractions which were present in the coolant in chemically combined forms. Table V shows the concentrations of the activation products as a function of reactor power.

3. Corrosion Products

Analyses were performed on aliquots of the same samples and on the separated crud fractions for important radionuclides resulting from neutron activation of corrosion products. The concentrations of these species as a function of reactor power are shown in Table VI. At reactor power levels less than 38.9% of full power, Mn^{56} was the only activated corrosion product detected in either the aqueous or solid phases. Figure 8 shows a portion of the Mn^{56} spectrum obtained from the chemically separated manganese fraction at 77.6% of full power.

The first indication of a significant quantity of another activated corrosion product was the appearance of a 0.51-Mev photopeak in the separated crud fraction obtained at 38.9% of full power. A component analysis of the decay curve for this photopeak yielded two distinct components having half-lives of approximately 12.5 hours and 15 to 20 minutes, respectively. A typical decay curve and component analysis is shown in Figure 9. The half-life and gamma energy of the longer lived component indicated that 12.8 hour Cu^{64} might be the source of this radiation. Radiochemical analysis of the dissolved crud sample obtained at 53% of full power confirmed the presence of Cu^{64} and yielded a quantitative value for its concentration. At this power level, Cu^{64} contributed less than 10% of the total gamma radioactivity in the crud fraction.

Analyses were also performed in both the aqueous and solid phases for Cr^{51}, Mn^{54}, Fe^{59}, $Co^{58, 60}$, Zr^{95}, and W^{187}. The analytical data are shown in Table VI. The concentrations of most of these nuclides in each phase were below the limits of detection of the analytical methods utilized for analyses performed at most reactor power levels. In those instances where definitive concentrations of such species as Co^{58}, Co^{60}, Cr^{51}, and Mn^{54} were observed, sample volumes in excess of 10 liters were taken for analysis.

TABLE V

CONCENTRATIONS OF ACTIVATION PRODUCTS
IN THE PRIMARY COOLANT[a]

Reactor Power (% of Full Power)	N^{13} Concentration [c] (dpm/ml)	(μc/ml)	F^{18} Concentration [b] (dpm/ml)	(μc/ml)	Na^{24} Concentration [b] (dpm/ml)	μc/ml)
9.7	8.7×10^3	3.9×10^{-3}	2.8×10^3	1.3×10^{-3}	3.2×10^2	1.4×10^{-4}
18.0	1.6×10^4	7.2×10^{-3}	7.0×10^3	3.2×10^{-3}	9.9×10^2	4.4×10^{-4}
38.9	8.5×10^4	3.8×10^{-2}	1.5×10^4	6.8×10^{-2}	3.1×10^3	1.4×10^{-3}
53.0	9.5×10^4	4.2×10^{-2}	1.9×10^4	8.5×10^{-3}	5.9×10^3	2.7×10^{-3}
77.6	1.6×10^5	7.2×10^{-2}	2.0×10^4	9.0×10^{-3}	7.8×10^3	3.5×10^{-3}
88.7	2.0×10^5	9.0×10^{-2}	3.3×10^4	1.5×10^{-2}	1.1×10^4	5.0×10^{-3}

(a) Data corrected for decay to time of sampling.

(b) Concentrations determined by radiochemical analyses.

(c) Concentrations determined by component analysis of integral decay curve of filtered and

degassed coolant sample.

TABLE VI

CONCENTRATIONS OF ACTIVATED CORROSION PRODUCTS IN THE PRIMARY COOLANT [a][b]

CRUD FRACTION

Reactor Power (% of Full Power)	Mn^{56} (dpm/ml)	Mn^{56} ($\mu c/ml$)	Cu^{64} (dpm/ml)	Cu^{64} ($\mu c/ml$)	$Co^{58, 60}$ (dpm/ml)	$Co^{58, 60}$ ($\mu c/ml$)	Fe^{59} (dpm/ml)	Fe^{59} ($\mu c/ml$)	Cr^{51} (dpm/ml)	Cr^{51} ($\mu c/ml$)	Mn^{54} (dpm/ml)	Mn^{54} ($\mu c/ml$)	Zr^{95} (dpm/ml)	Zr^{95} ($\mu c/ml$)
4.5	4.5×10^{1}	2.0×10^{-5}	N.D.[d]	-	-	-	-	-	-	-	-	-	-	-
9.7	2.7×10^{2}	1.2×10^{-4}	N.D.	-	N.D.	-	N.D.	-	N.D.	-	N.D.	-	-	-
18.0	8.0×10^{2}	3.6×10^{-4}	N.D.	-	N.D.	-	N.D.	-	N.D.	-	N.D.	-	-	-
38.9	2.2×10^{2}	1.0×10^{-4}	3.7×10^{1}	1.7×10^{-5}	N.D.	-	N.D.	-	N.D.	-	N.D.	-	N.D.	-
53.0	7.6×10^{2}	3.9×10^{-4}	5.2×10^{1}	2.3×10^{-5}	-	-	-	-	-	-	-	-	-	-
77.6	6.5×10^{2}	2.9×10^{-4}	7.8×10^{1}	3.5×10^{-5}	3.7	1.7×10^{-6}	1.2	5.4×10^{-7}	1.4	6.3×10^{-7}	2.6×10^{-1}	1.2×10^{-7}	-	-
88.7	1.2×10^{3}	5.4×10^{-4}	1.1×10^{2}	5.0×10^{-5}	5.0	2.3×10^{-6}	1.4	6.3×10^{-7}	1.0	4.5×10^{-7}	3.6×10^{-1}	1.6×10^{-7}	-	-
100.0 [c]	-	-	-	-	1.3×10^{1}	5.9×10^{-6}	3.1	1.4×10^{-6}	3.9	1.8×10^{-6}	9.1×10^{-1}	4.1×10^{-7}	1.2	5.4×10^{-7}

AQUEOUS FRACTION

Reactor Power (% of Full Power)	Mn^{56} (dpm/ml)	Mn^{56} ($\mu c/ml$)	Cu^{64} (dpm/ml)	Cu^{64} ($\mu c/ml$)	$Co^{58, 60}$ (dpm/ml)	$Co^{58, 60}$ ($\mu c/ml$)	Fe^{59} (dpm/ml)	Fe^{59} ($\mu c/ml$)	Cr^{51} (dpm/ml)	Cr^{51} ($\mu c/ml$)	Mn^{54} (dpm/ml)	Mn^{54} ($\mu c/ml$)	Zr^{95} (dpm/ml)	Zr^{95} ($\mu c/ml$)
9.7	3.5×10^{3}	1.6×10^{-3}	-	-	-	-	-	-	-	-	-	-	-	-
18.0	1.3×10^{4}	5.9×10^{-3}	-	-	N.D.	-	N.D.	-	N.D.	-	N.D.	-	-	-
38.9	2.9×10^{4}	1.3×10^{-2}	-	-	N.D.	-	N.D.	-	N.D.	-	N.D.	-	-	-
53.0	1.2×10^{5}	5.4×10^{-2}	N.D.	-	1.1	5.0×10^{-7}	7.8×10^{-1}	3.5×10^{-7}	2.0×10^{-1}	9.0×10^{-8}	2.5×10^{1}	1.1×10^{-5}	1.2×10^{-1}	5.4×10^{-2}
77.6	7.9×10^{4}	3.6×10^{-2}	N.D.	-	-	-	-	-	-	-	-	-	N.D.	-
88.7	1.1×10^{5}	5.0×10^{-2}	-	-	-	-	-	-	-	-	-	-	-	-
100.0 [c]	-	-	-	-	7.0×10^{-2}	3.2×10^{-8}	8.0×10^{-1}	3.6×10^{-7}	2.4×10^{-1}	1.1×10^{-7}	2.5×10^{1}	1.1×10^{-5}	1.6×10^{-1}	7.2×10^{-8}

(a) All data corrected to time of sampling.
(b) No detectable concentrations of W^{187} were observed in either fraction at any power level cited.
(c) Special sample obtained for determination of long-lived activated corrosion products. Power level had been at 100% followed by sharp transient to 40%.
(d) N.D. = Not detected.

FIGURE 8

Typical gamma spectrum (0 to 1.4 Mev) of separated manganese fraction. Reactor at 77.6% of full power
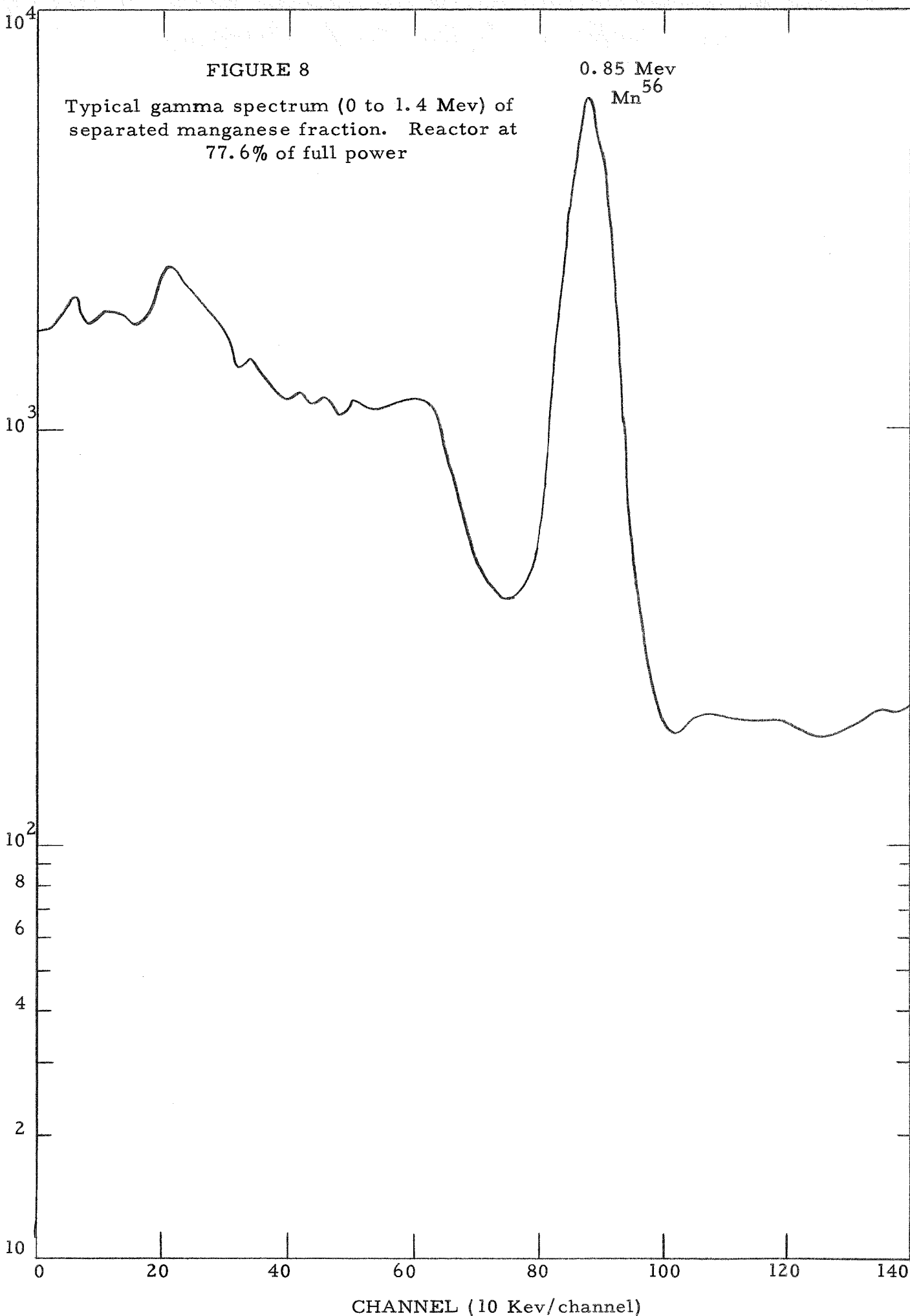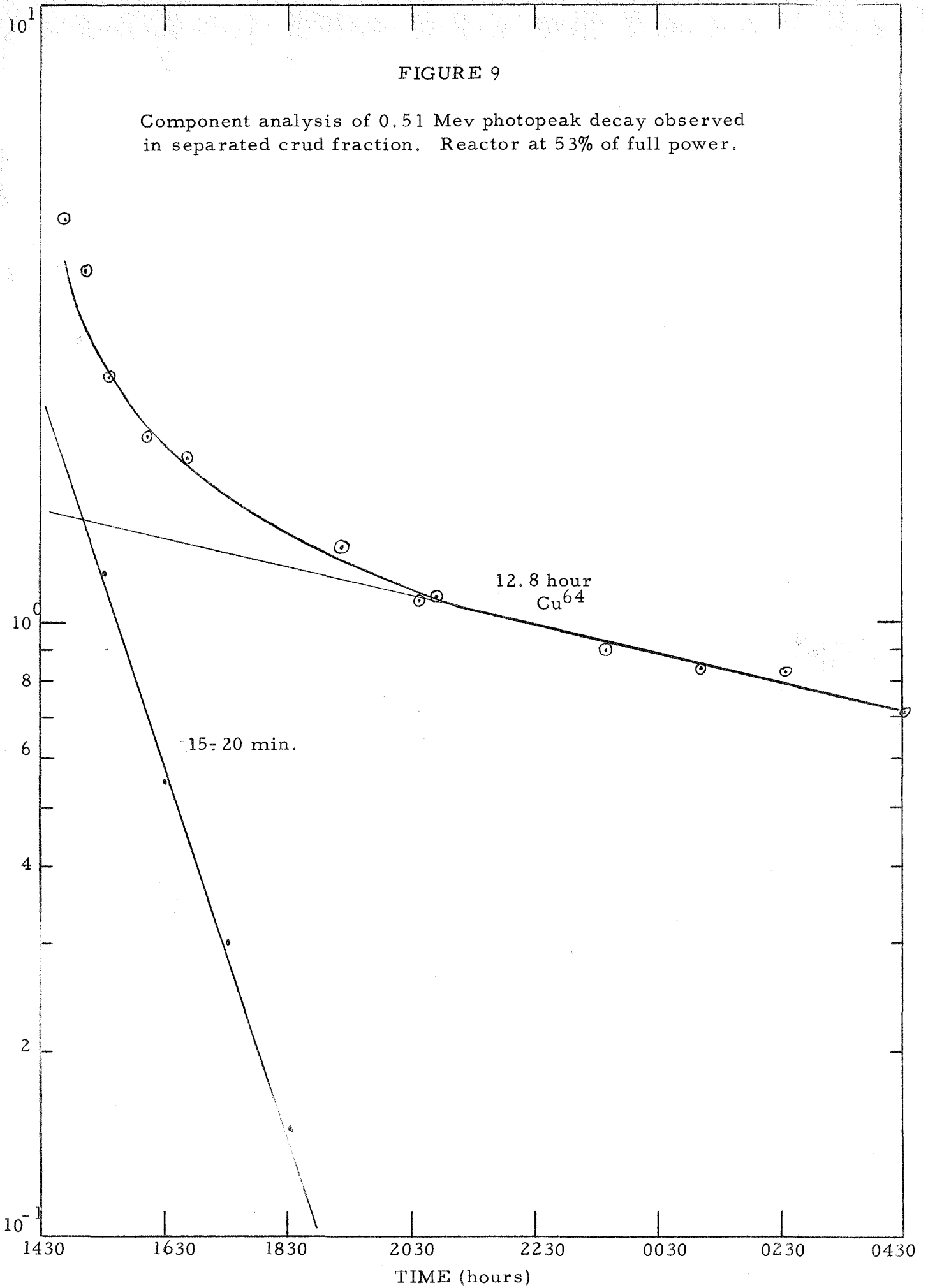
0.85 Mev
Mn^{56}

CHANNEL (10 Kev/channel)

COUNTS/CHANNEL/2MIN.

FIGURE 9

Component analysis of 0.51 Mev photopeak decay observed in separated crud fraction. Reactor at 53% of full power.

12.8 hour Cu^{64}

15-20 min.

- 29 -

4. Fission Products

The important fission products selected for investigation included $I^{131, 133, 134, 135}$, Cs^{138}, $Sr^{89, 90, 91, 92}$, and $Ba^{139, 140}$. Analyses were performed for these nuclides in the crud and/or aqueous fractions. All data except those for $Sr^{89, 90}$ were obtained by gamma spectrometry measurements and integral gamma countings on separated fractions of the respective elements. The beta radiation of $Sr^{89, 90}$ were measured with end-window, methane-flow proportional counters.

Two methods of data treatment were used to determine the concentrations of the four iodine isotopes: (a) gamma spectrometry, and (b) component analysis of the integral decay curve of the mixed iodine radioactivities. All four isotopes were determined by the first method, while the values for I^{131} and I^{134} were confirmed by the second method. Figure 10 shows a typical gamma spectrum of an iodine sample shortly after radiochemical purification, and Figure 11 is a spectrum of the same sample at a later time showing the decay of the iodine isotopes and the growth of the Xe^{133} and Xe^{135m} daughters of I^{133} and I^{135}. Figure 12 illustrates the decay curves of the individual photopeaks shown in Figures 10 and 11. A typical integral decay curve for the mixed iodine radioactivities, illustrating the method of component analysis, has been presented in Figure 2. The concentration data for these radio-nuclides are shown in Table VII as a function of reactor power.

The 32-minute Cs^{138} was the only cesium isotope observed in the primary coolant during this program. It was determined by measurement of its half-life and gamma energy. Gamma spectra of the separated cesium fractions exhibited no other gamma radiations, and no evidence of a longer lived cesium isotope existed in the integral decay curves. A typical spectrum of the separated cesium fraction is shown in Figure 13, and a typical integral decay curve in Figure 14. The data for Cs^{138} concentrations as a function of reactor power are shown in Table VIII.

The 9.5-minute Cs^{139} is normally expected to be present in PWR type primary coolants along with Cs^{138}. Measurement of this nuclide was precluded by the relatively long flow time of the primary coolant in the sampling line and the normal time requirement for separation of the cesium fraction.

The strontium isotopes, 9.2-hour Sr^{91} and 2.7-hour Sr^{92}, were determined in the primary coolant by gamma spectrometry measurements of the separated

FIGURE 10

Typical gamma spectrum of separated iodine fraction 63 minutes after separation. Reactor at 88.7% of full power.

FIGURE 11
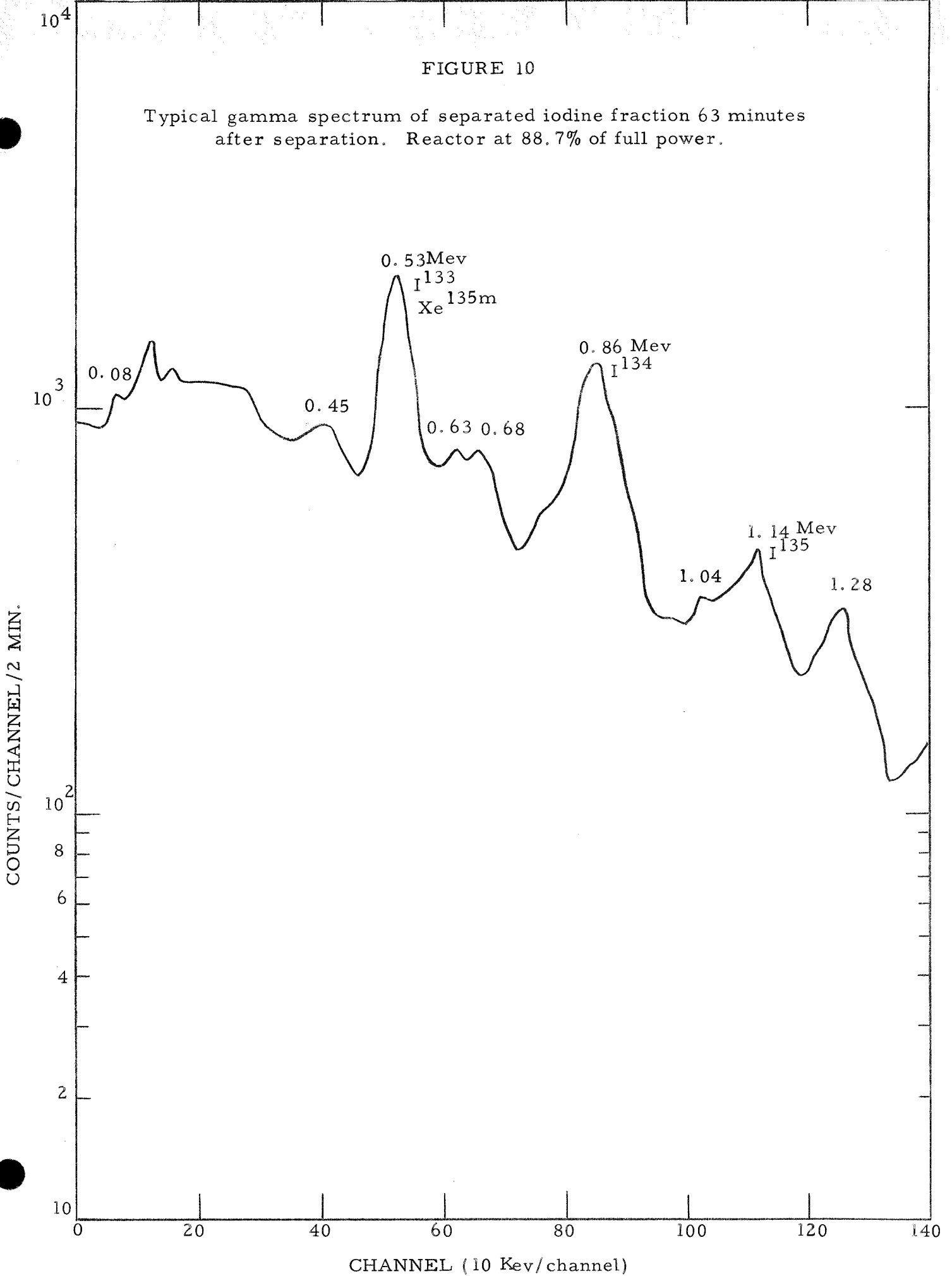
Typical gamma spectrum of separated iodine fraction 14.5 hours after separation. Reactor at 88.7% of full power.
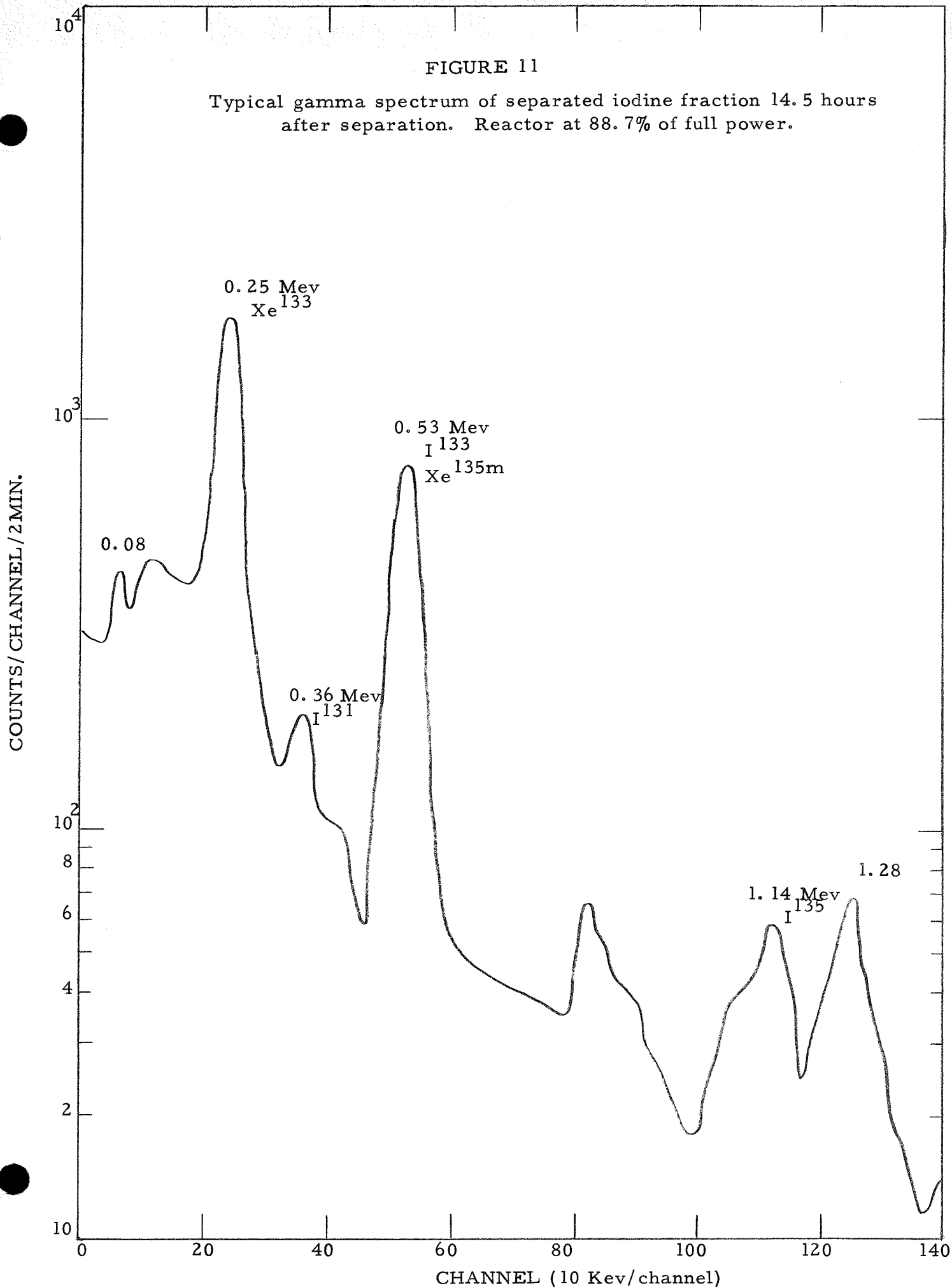
- 32 -

FIGURE 12

Decay curves of characteristic photopeaks of I$^{131, 133, 134, 135}$.
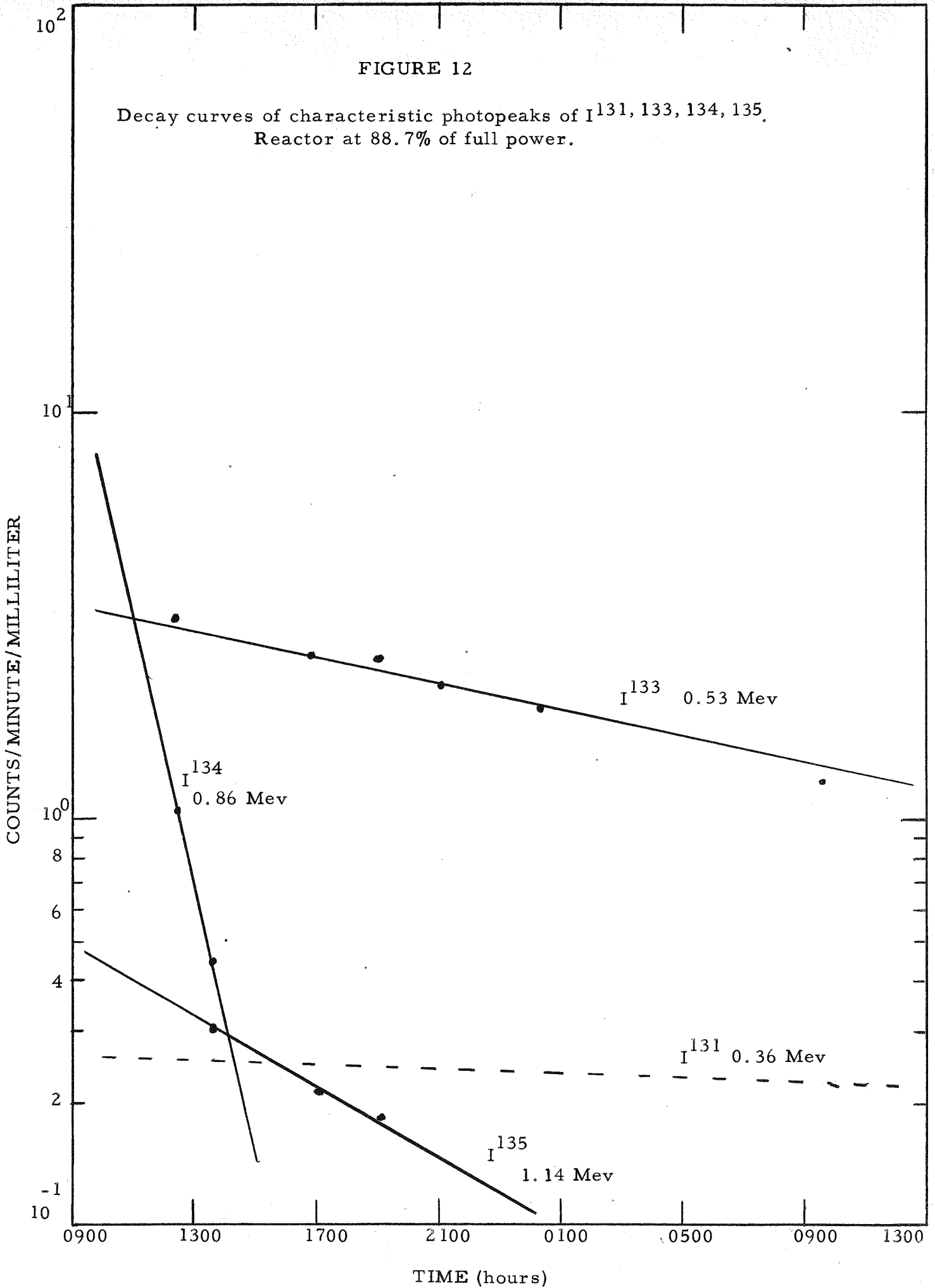### Reactor at 88.7% of full power.

TABLE VII

CONCENTRATIONS OF IODINE ISOTOPES IN THE PRIMARY COOLANT

Nuclide	Reactor Power (% of Full Power)	Concentration (dpm/ml)	(μc/ml)
I^{131} (a)	9.7	N.D.	----
(a) (b)	18.0	N.D.	----
(a) (b)	38.9	N.D.	----
(a) (b)	53.0	N.D.	----
(a)	77.6	1.0×10^{1}	4.5×10^{-6}
(b)		3.3	1.5×10^{-6}
(a)	88.7	1.1×10^{1}	5.0×10^{-6}
(b)		4.7	2.1×10^{-6}
I^{133} (b)	9.7	7.0	3.2×10^{-6}
	18.0	1.7×10^{1}	7.7×10^{-6}
	38.9	2.4×10^{1}	1.1×10^{-5}
	53.0	1.6×10^{1}	7.2×10^{-6}
	77.6	4.6×10^{1}	2.1×10^{-5}
	88.7	7.5×10^{1}	3.4×10^{-5}

Nuclide	Reactor Power (% of Full Power)	Concentration (dpm/ml)	(μc/ml)
I^{134} (a)	9.7	3.8×10^{1}	1.7×10^{-5}
(a)	18.0	5.8×10^{1}	2.6×10^{-5}
(b)		7.6×10^{1}	3.4×10^{-5}
(a)	38.9	1.6×10^{2}	7.2×10^{-5}
(b)		1.7×10^{2}	7.7×10^{-5}
(a)	53.0	2.1×10^{2}	9.5×10^{-5}
(b)		1.3×10^{2}	5.9×10^{-5}
(a)	77.6	3.2×10^{2}	1.4×10^{-4}
(b)		2.5×10^{2}	1.1×10^{-4}
(a)	88.7	4.0×10^{2}	1.8×10^{-4}
(b)		3.7×10^{2}	1.7×10^{-4}
I^{135} (b)	9.7	N.D.	-----
	18.0	N.D.	-----
	38.9	6.9×10^{1}	3.1×10^{-5}
	53.0	4.6×10^{1}	2.1×10^{-5}
	77.6	7.0×10^{1}	3.2×10^{-5}
	88.7	9.7×10^{1}	4.4×10^{-5}

(a) Data obtained from integral gamma decay of separated iodine fraction.

(b) Data obtained from gamma spectral analysis of separated iodine fraction.

FIGURE 13

Typical spectrum of separated cesium fraction.
Reactor at 88.7% of full power.

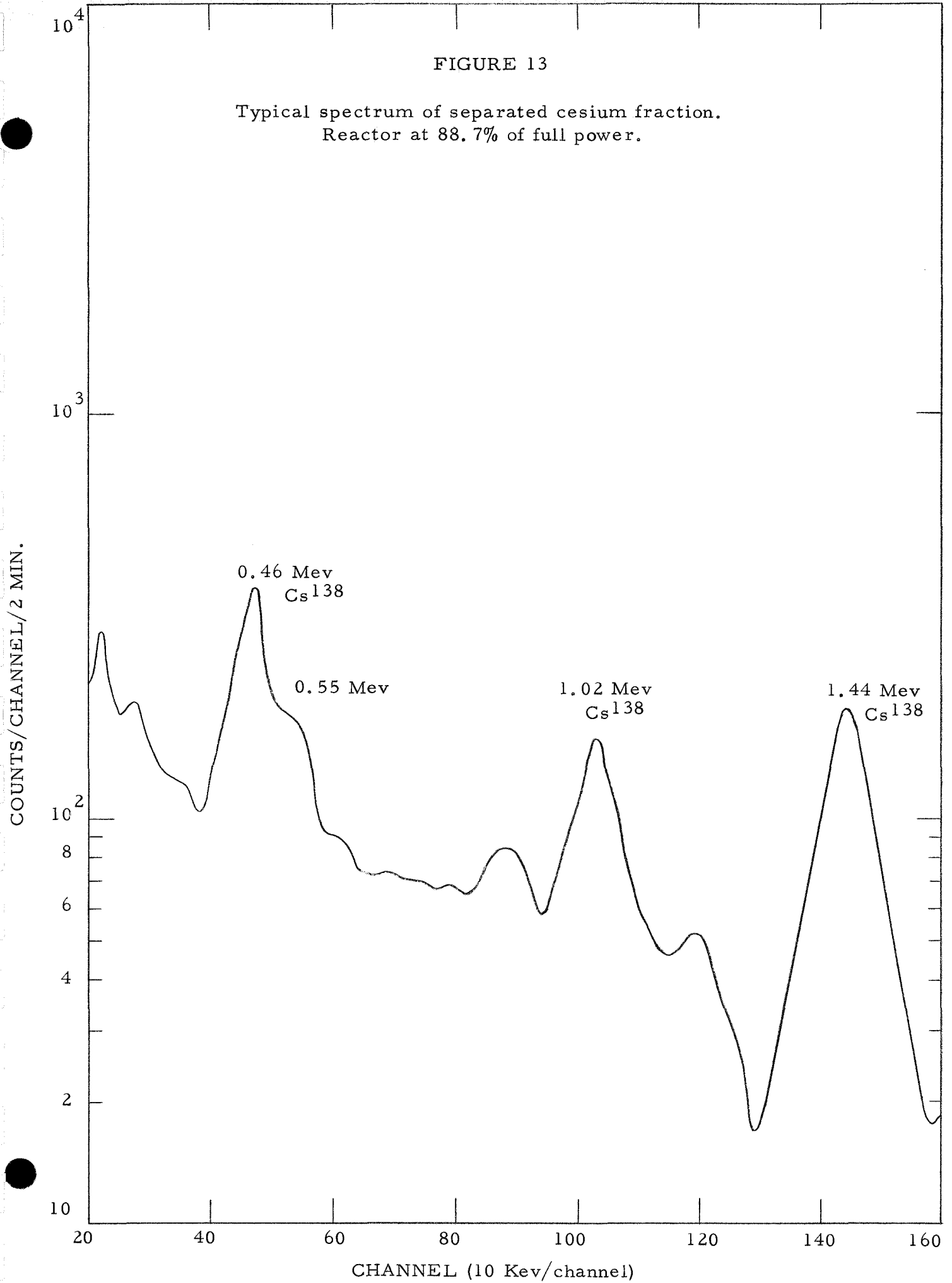0.46 Mev
Cs^{138}

0.55 Mev

1.02 Mev
Cs^{138}

1.44 Mev
Cs^{138}

CHANNEL (10 Kev/channel)

FIGURE 14

Integral decay curve separated cesium fraction. Reactor at 38.9% of full power.
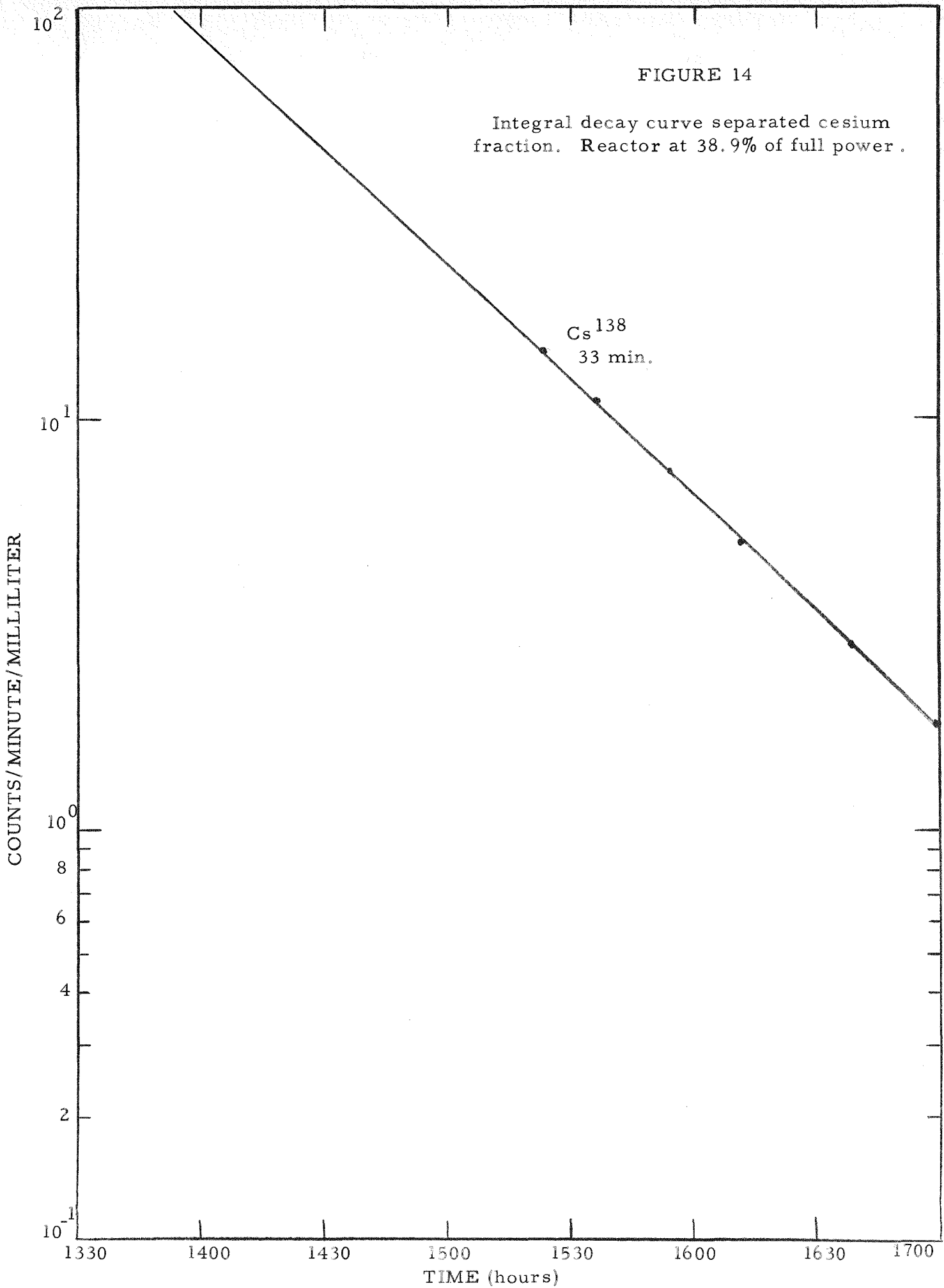
Cs^{138}
33 min.

COUNTS/MINUTE/MILLILITER

TIME (hours)

TABLE VIII

CONCENTRATIONS OF Cs^{138} IN THE PRIMARY COOLANT

Reactor Power (% of Full Power)	Concentration (dpm/ml) *	Concentration (μc/ml) *
9.7	3.1×10^{1}	1.4×10^{-5}
18.0	5.8×10^{1}	2.6×10^{-5}
38.9	1.4×10^{2}	6.1×10^{-5}
53.0	1.8×10^{2}	8.0×10^{-5}
77.6	2.9×10^{2}	1.3×10^{-4}
88.7	3.4×10^{2}	1.5×10^{-4}

* Corrected to time of sampling.

strontium fractions. Figures 15 and 16 show typical spectra of a separated strontium fraction as a function of decay time. The concentration data as a function of reactor power are shown in Table IX.

Analyses for Sr^{91} and Sr^{92} were also performed on separated crud fractions at power levels above 18% of full power. No significant concentration was observed in these samples.

Analyses for Sr^{89} and Sr^{90} were performed on primary coolant and separated crud fractions at 9.7%, 18%, 53% and 100% of full power. No Sr^{89} or Sr^{90} activity was observed in any crud fraction. These two species were first detected in the primary coolant sample obtained at 53% of full power, in which their respective concentrations were $1.1 \times 10^{-8} \mu c/ml$, and $1.1 \times 10^{-9} \mu c/ml$. The observed concentrations at 100% of full power were $5.9 \times 10^{-8} \mu c/ml$ and $1.3 \times 10^{-9} \mu c/ml$, respectively.

Analyses of primary coolant were performed for Ba^{139} and Ba^{140} at the power levels cited in Table IX. A typical gamma spectrum of a separated barium fraction is shown in Figure 17. The prominent 0.165-Mev photopeak of the Ba^{139} is clearly visible. Indications of photopeaks of Ba^{140} are present, although they are too small for use in quantitative measurements. Therefore, the Ba^{139} and Ba^{140} were determined by component analysis of the integral decay curve of the barium fraction. A typical decay curve is shown in Figure 18. Values were first obtained for Ba^{139} at 18% of full power. Barium-140 was detected initially at 38.9% of full power. The values of the concentrations of these nuclides as a function of reactor power are shown in Table IX. No barium activity was observed in any crud fraction analyzed during this program.

Four fission products, I^{134}, I^{133}, Cs^{138}, and Ba^{139}, were selected for use in estimating the uranium contamination on the reactor core surfaces. The observed values of the concentrations of these fission products were determined at steady-state reactor operations. After normalization to 100% of full power, the mean values were used to compute the uranium content of the core surfaces using the method described in the appendix. The data are presented in the next section.

FIGURE 15

Typical spectrum of separated strontium fraction 20 minutes after separation. Reactor at 88.7% of full power.
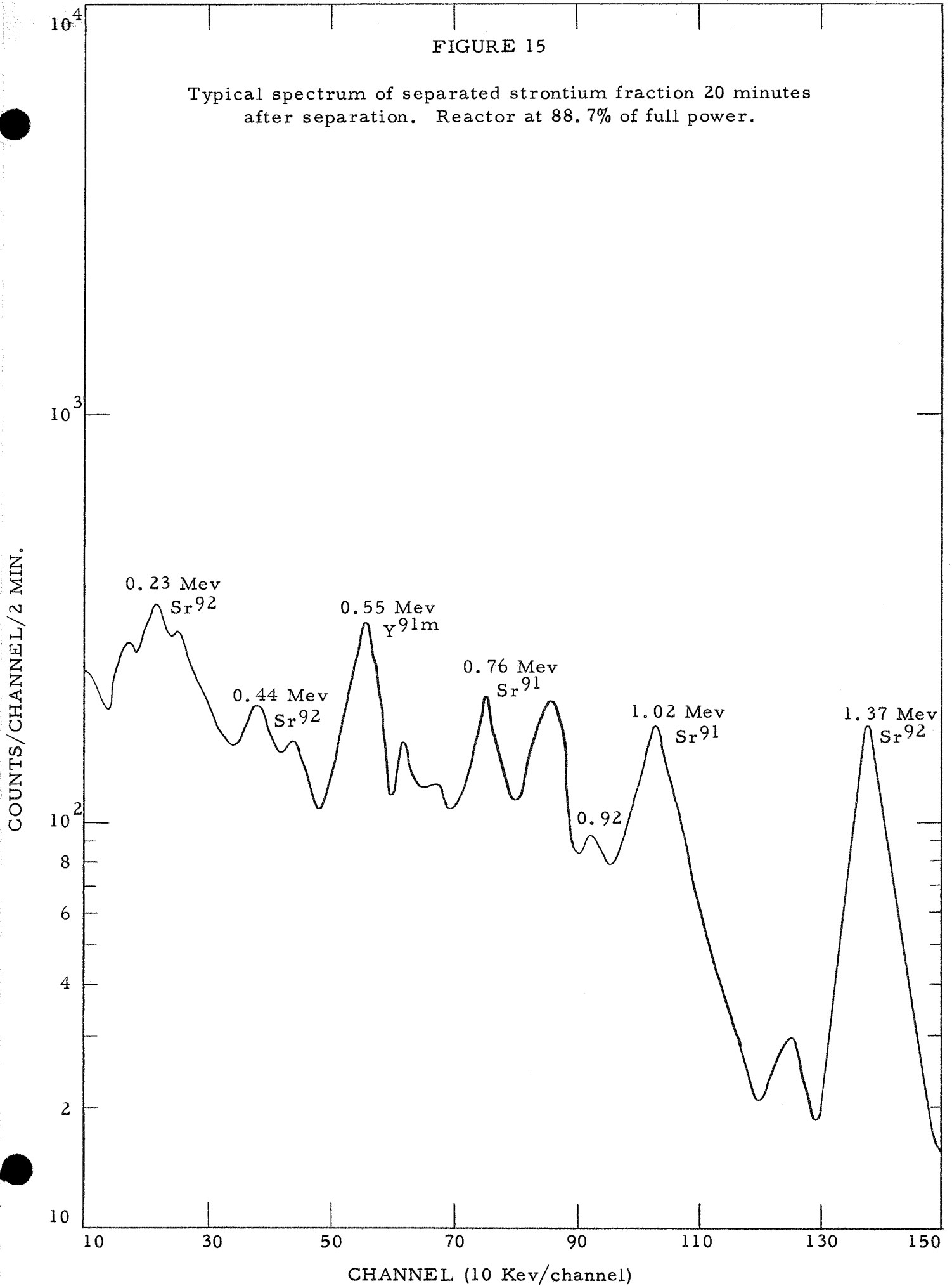
FIGURE 16

Typical spectrum of separated strontium fraction 6.2 hours after separation. Reactor at 88.7% of full power.
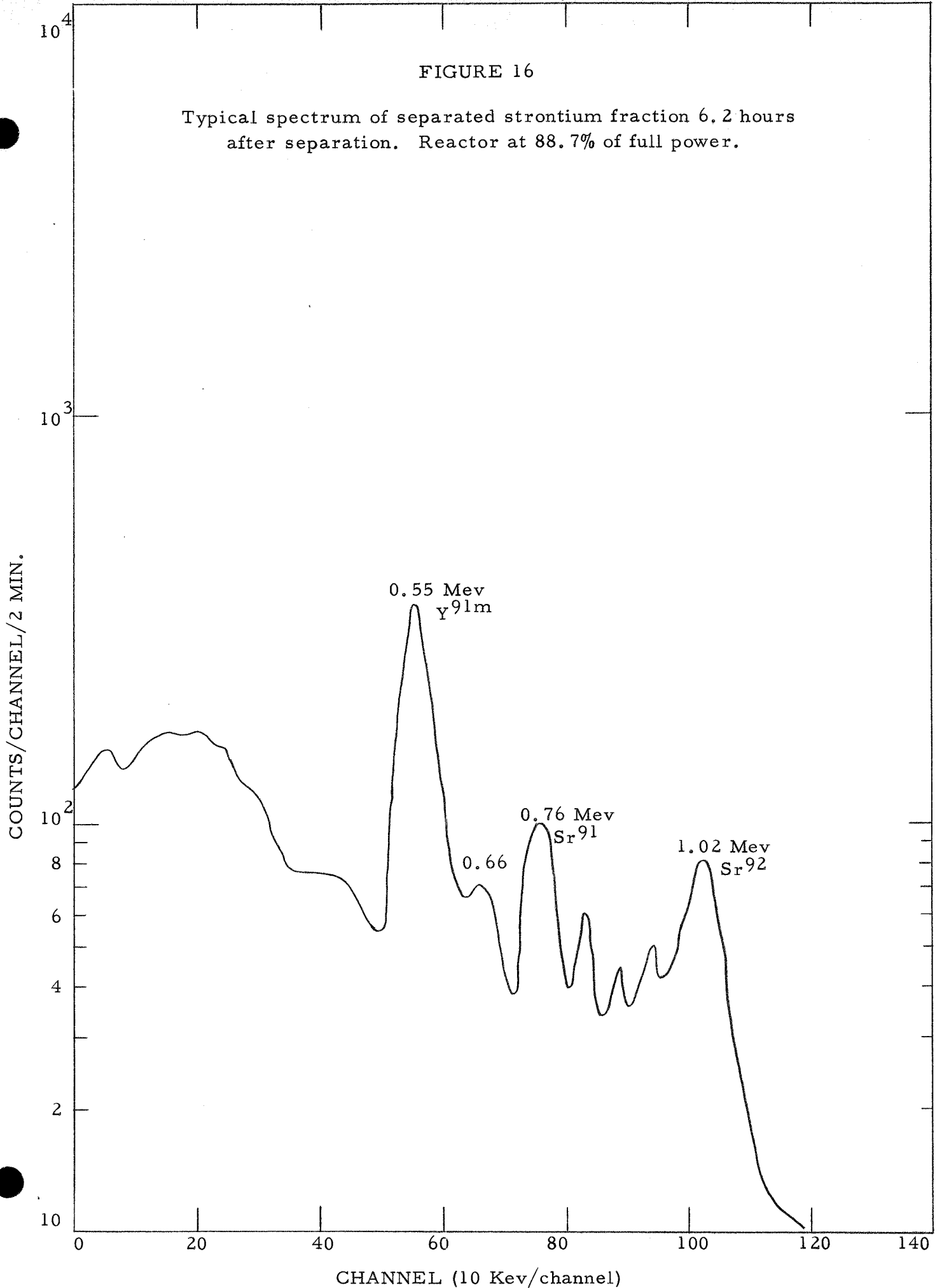
CHANNEL (10 Kev/channel)

TABLE IX

CONCENTRATIONS OF STRONTIUM AND BARIUM ISOTOPES IN THE AQUEOUS FRACTION OF THE PRIMARY COOLANT[a]

Reactor Power (% of Full Power)	Sr^{91} [b] (dpm/ml)	(μc/ml)	Sr^{92} [b] (dpm/ml)	(μc/ml)	Ba^{139} [c] (dpm/ml)	(μc/ml)	Ba^{140} [c] (dpm/ml)	(μc/ml)
9.7	N.D.	-	N.D.	-	N.D.	-	N.D.	-
18.0	N.D.	-	N.D.	-	1.8×10^1	7.9×10^{-6}	N.D.	-
38.9	6.0×10^1	2.7×10^{-5}	1.2×10^2	5.0×10^{-5}	9.0×10^1	4.1×10^{-5}	1.8	8.1×10^{-7}
53.0	3.1×10^1	1.4×10^{-5}	9.3×10^1	4.2×10^{-5}	8.1×10^1	3.7×10^{-5}	2.9	1.3×10^{-6}
77.6	3.4×10^1	1.5×10^{-5}	6.1×10^1	2.6×10^{-5}	2.0×10^2	9.1×10^{-5}	4.2	1.9×10^{-6}
88.7	4.1×10^1	1.8×10^{-5}	9.0×10^1	4.1×10^{-5}	1.6×10^2	7.2×10^{-5}	4.8	2.2×10^{-6}

(a) No significant concentrations of Sr or Ba were found in the particulate fractions.

(b) Concentrations determined by photopeak decay obtained from gamma spectra of chemically separated fractions.

(c) Concentrations determined by component analysis of integral decay curves of chemically separated fractions.

FIGURE 17

Typical gamma spectrum of a separated barium
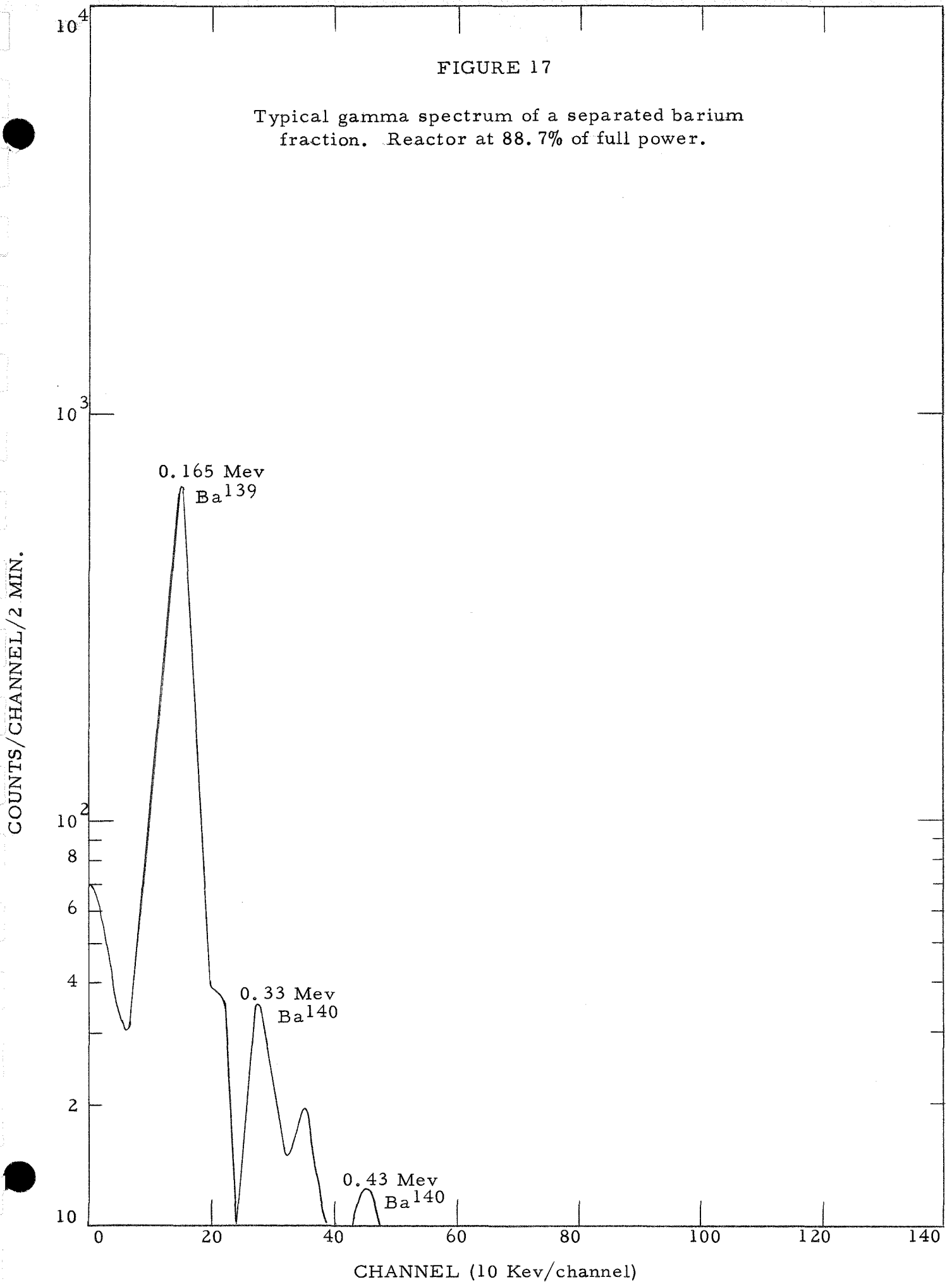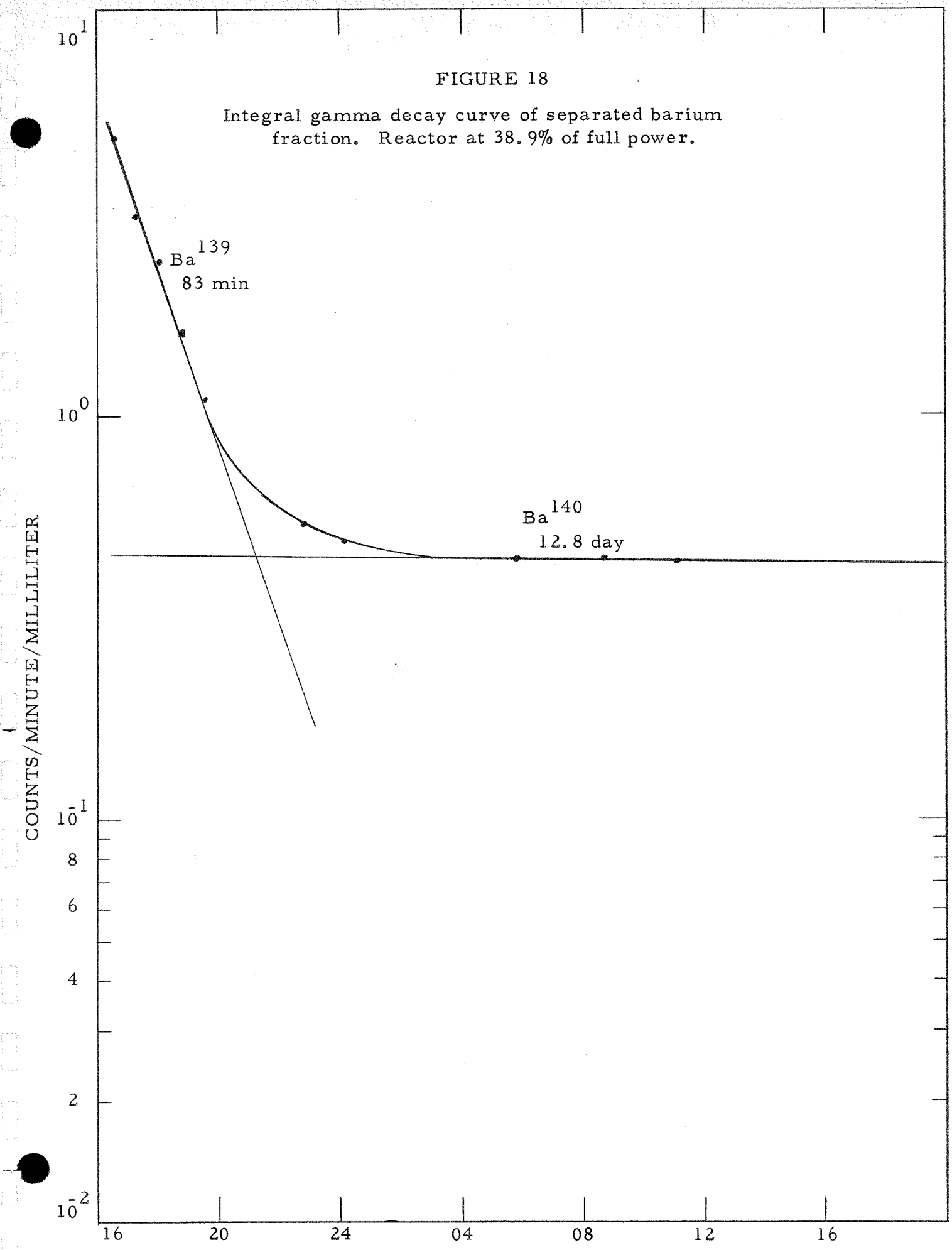fraction. Reactor at 88.7% of full power.

FIGURE 18

Integral gamma decay curve of separated barium
fraction. Reactor at 38.9% of full power.

- 43 -

C. Radioactivity Balance Studies

The detailed studies of specific radionuclides in the primary coolant at reactor power levels of 9.7%, 18%, 38.9%, 53%, 77.6%, and 88.6% of full power included treatment of the data to assure that all significant radioactive species in the coolant had been detected and measured. Comparison of the sums of the radioactivity concentrations of the chemically separated and purified fractions were compared to the concentrations of the gross gamma radioactivity observed in the same sample (one hour after sampling). The results of the radioactivity balances for the respective power levels are presented graphically in Figures 19 through 24.

The values of the individually determined species, corrected for decay to one hour after sampling, are tabulated in Tables X through XV. In each case, the value of the summation of the radioactivity concentrations was in agreement, within experimental error, with the value of the gross gamma radioactivity concentration. These results confirmed qualitative observations, made using gamma spectral data, indicating that all significant gamma-emitting nuclides had been studied. It may be seen, from the cited tables and figures, that Mn^{56} was the principal radioactive constituent of the coolant for periods on the order of one hour after sampling. Its contribution varied from 60% to 85% of the total radioactivity at each power level at this decay time. The fractional contributions of the majority of the radionuclides studied to the total gamma radioactivity remained fairly constant at the power levels at which studies were performed.

FIGURE 19

Radioactivity balance of primary coolant.
Reactor at 9.7% of full power.

- Filtered Degassed ▲ Gross Crud
- ⊙ Manganese ⊗ Strontium
- Fluorine △ Cesium
- ✕ Nitrogen

FIGURE 20

Radioactivity balance of primary coolant.
Reactor at 18% of full power.

- Filtered Degassed   ✕ Nitrogen
⊙ Manganese   ▲ Gross Crud
● Fluorine   ◖ Sodium
+ Iodine

- 46 -

FIGURE 21

Radioactivity balance of primary coolant.
Reactor at 38.9% of full power.

- Filtered Degassed   ◐ Sodium
⊙ Manganese   ▲ Gross Crud
✕ Nitrogen   △ Cesium
● Fluorine   ⊗ Strontium
+ Iodine

COUNTS/MINUTE/MILLILITER

TIME (hours)

- 47 -
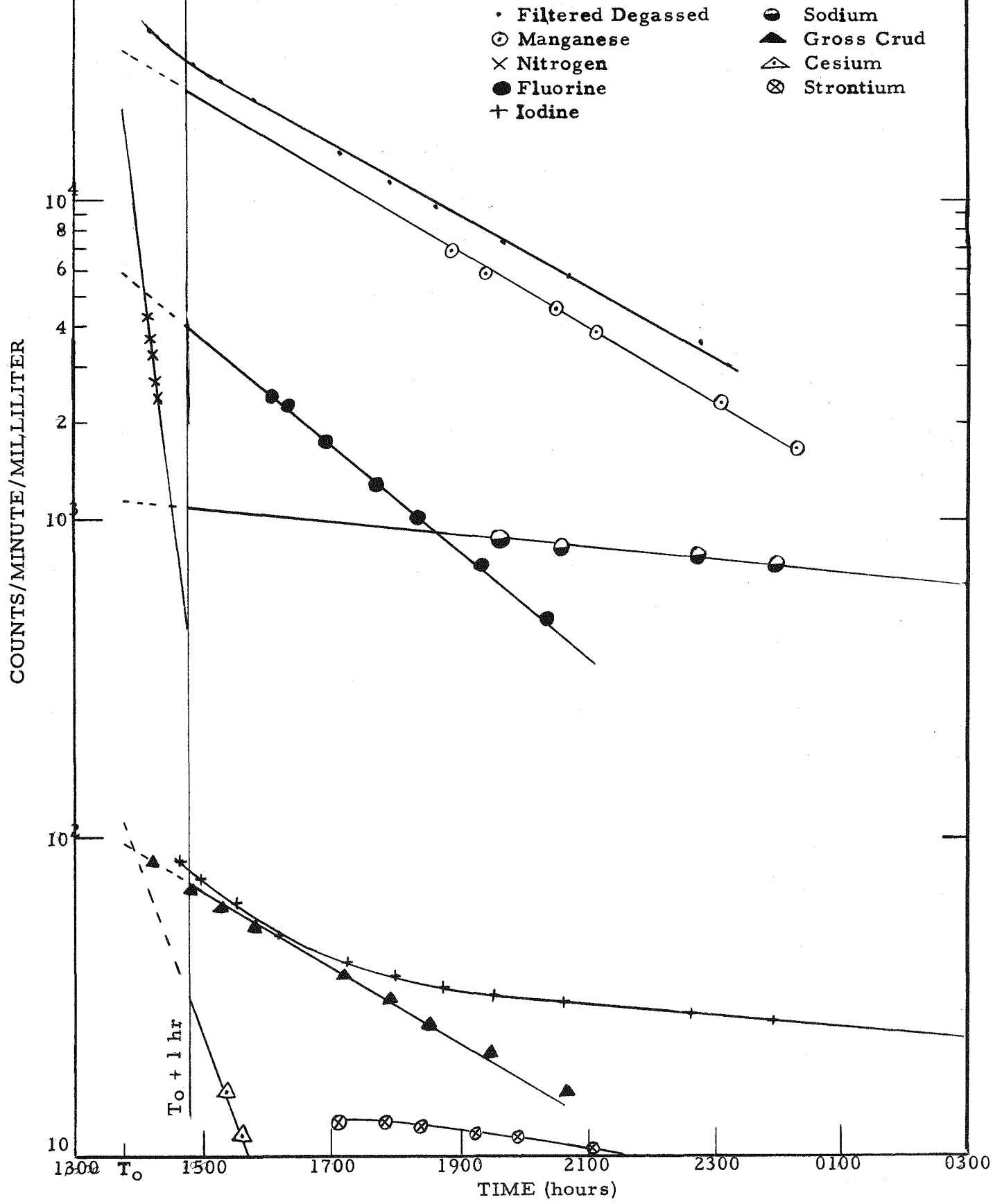
FIGURE 22

Radioactivity balance of primary coolant.
Reactor at 53% of full power.

⊗ Strontium   ⊖ Sodium
⊙ Manganese   ▲ Gross Crud
● Fluorine   △ Cesium
✕ Nitrogen   + Iodine
⊕ Degassed

COUNTS/MINUTE/MILLILITER

$T_o + 1$ hr

TIME (hours)

- 48 -

FIGURE 23

Radioactivity balance of primary coolant.
Reactor at 77.6% of full power.

- Filtered Degassed   ▲ Gross Crud
⊙ Manganese   △ Cesium
◑ Sodium   ⊗ Strontium
● Fluorine   ⊡ Barium
✕ Nitrogen   + Iodine
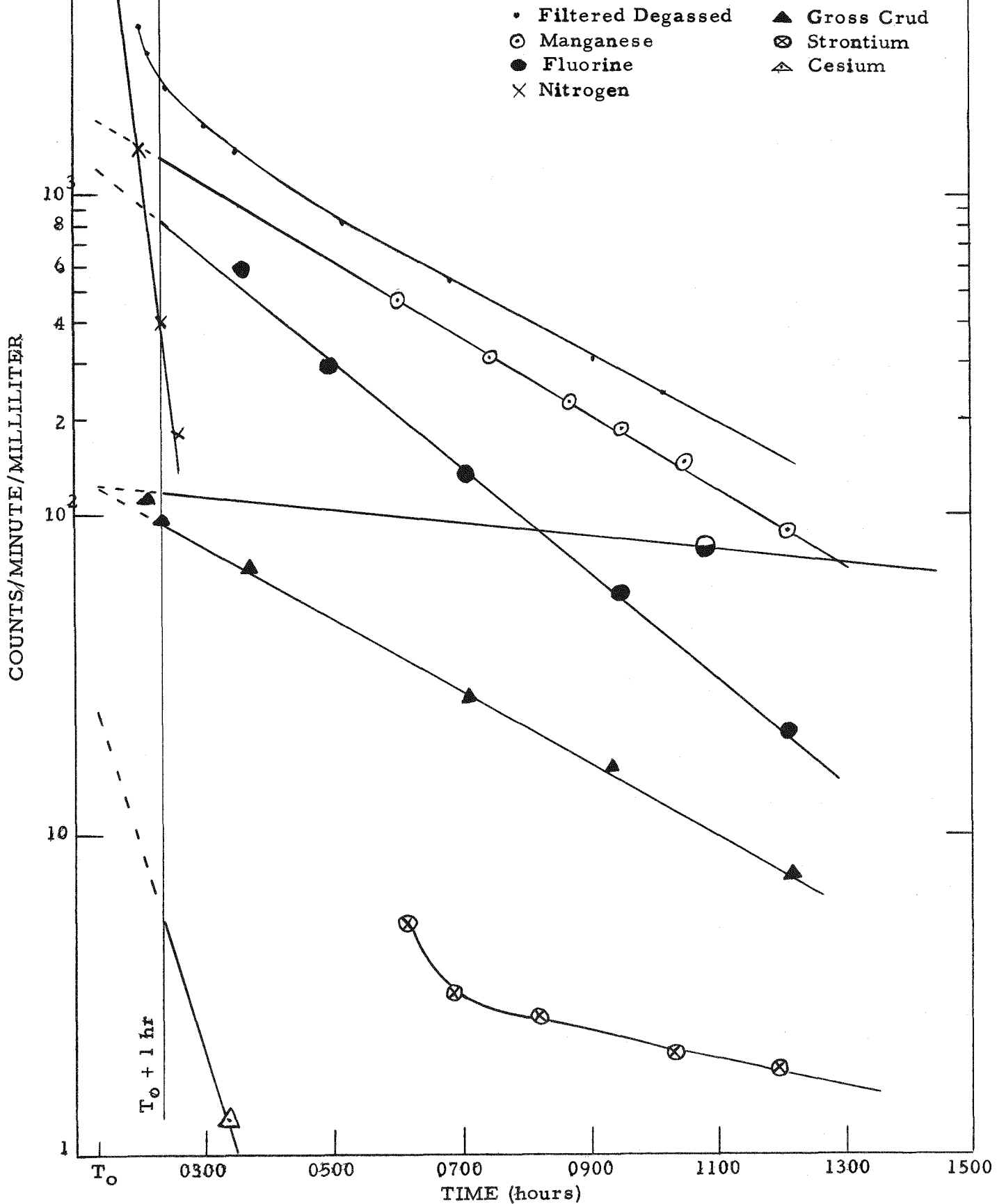
COUNTS/MINUTE/MILLILITER

TIME (hours)

- 49 -

FIGURE 24

Radioactivity balance of primary coolant.
Reactor at 88.7% of full power.

- Filtered Degassed
- ⊙ Manganese
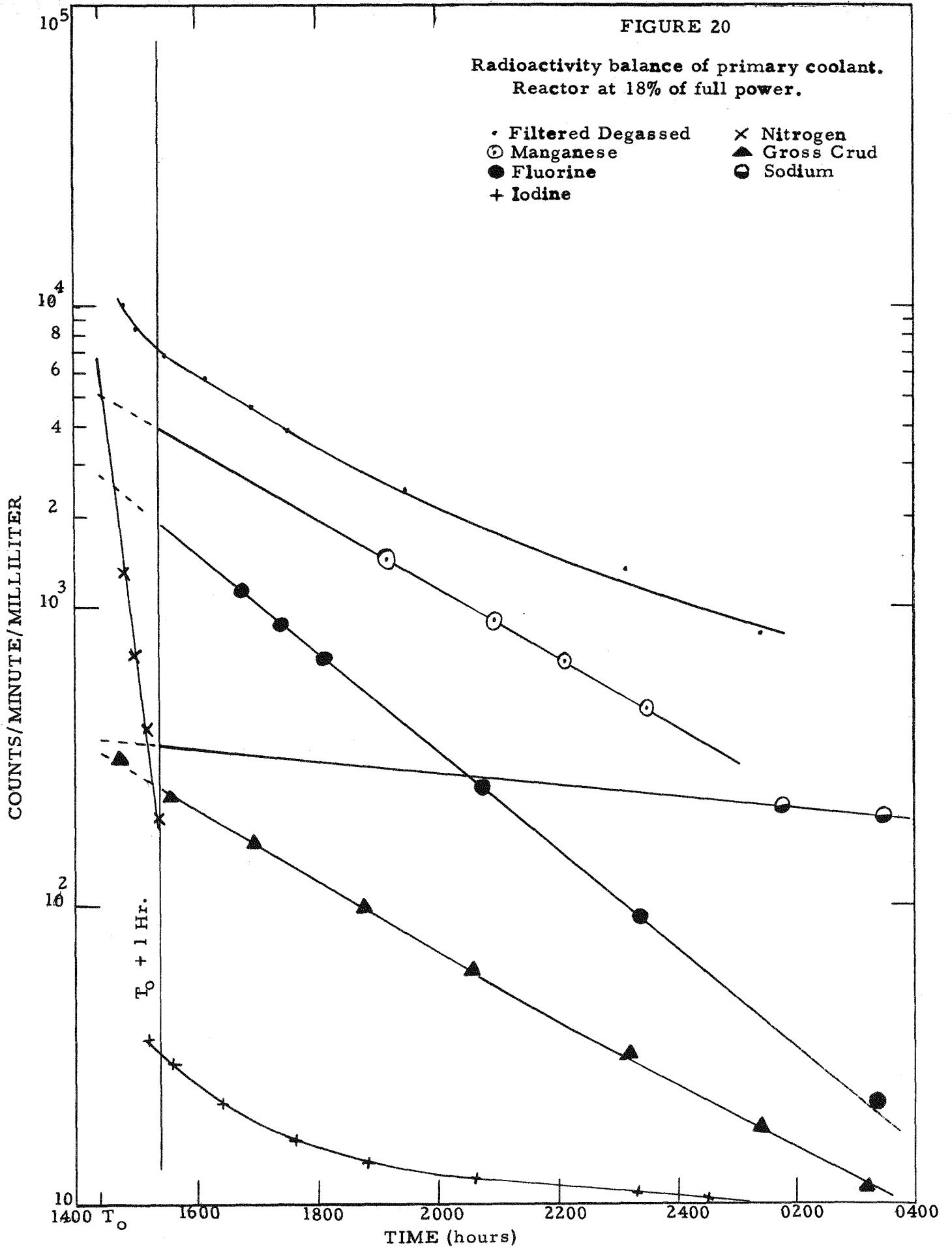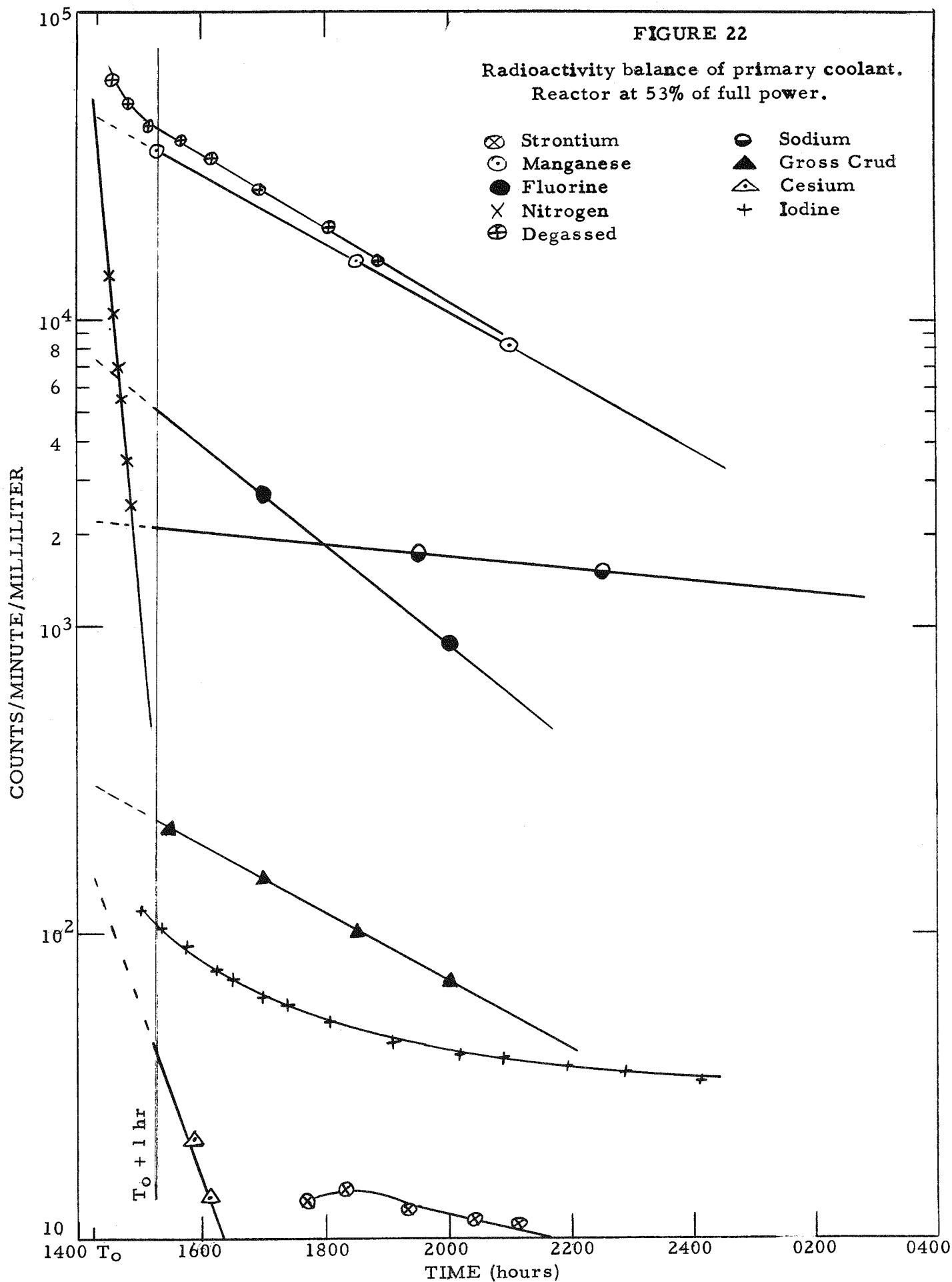- ⊖ Sodium
- ● Fluorine
- × Nitrogen
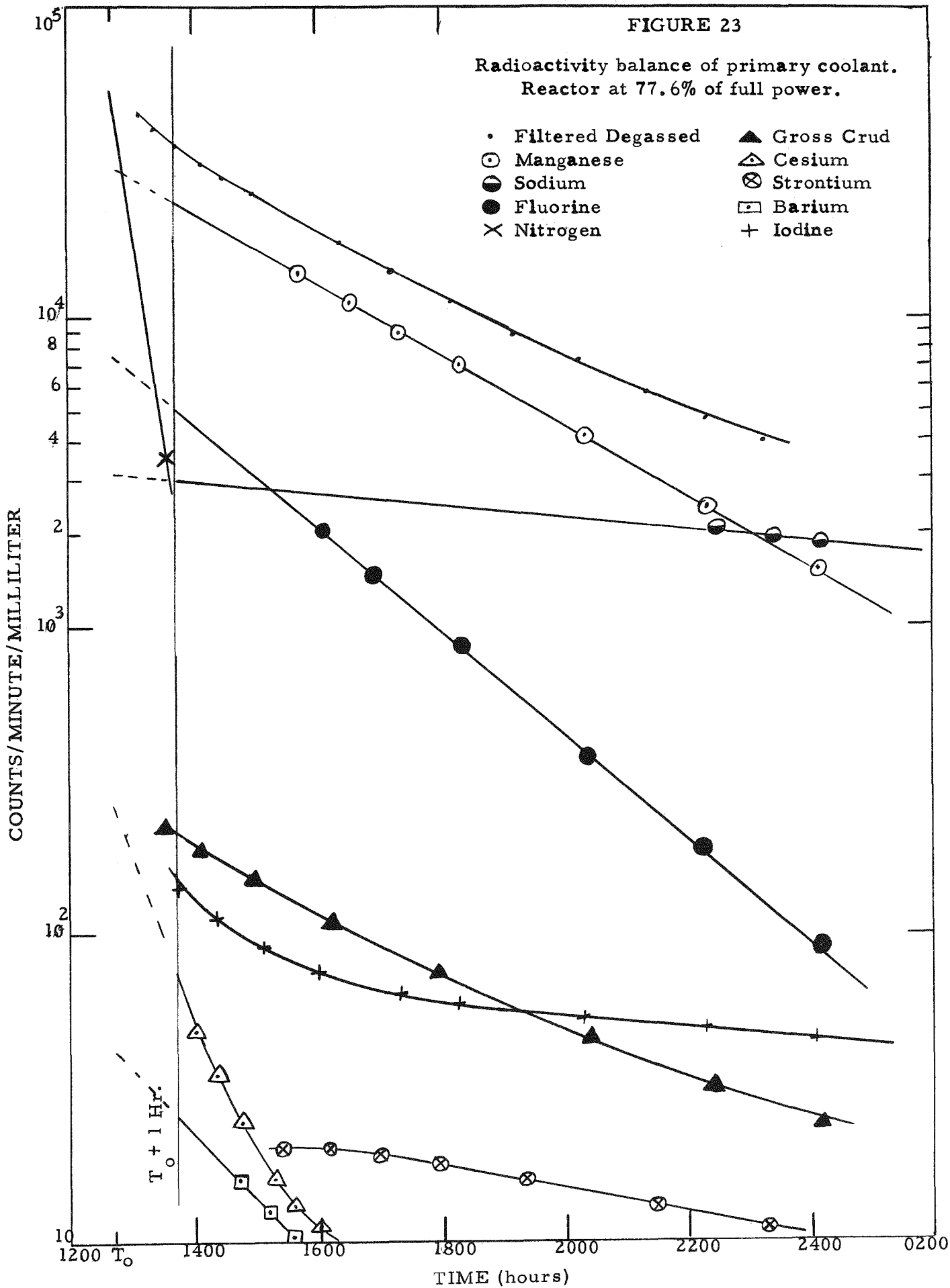- ▲ Gross Crud
- △ Cesium
- ⊕ Strontium
- ▣ Barium
- + Iodine

COUNTS/MINUTE/MILLILITER

TIME (hours)

TABLE X

RADIOACTIVITY BALANCE IN PRIMARY COOLANT

Date: 18 January, 1962   Reactor Power: 9.7%

Time: 0144 hours   Time at Power: 300 minutes

Sample Measured	Concentration * (cpm/ml)	Percentage of Total
Gross filtered degassed	1.9×10^3	--
Mn^{56}	1.1×10^3	57.5
F^{18}	7.5×10^2	38.2
Na^{24}	1.1×10^2	5.8
N^{13} (non-degassable)	1.6×10^1	0.8
Gross I	6.5×10^0	0.3
Gross Cs	2.5×10^1	1.3
		103.9

*Corrected to a decay time of one hour after sampling.
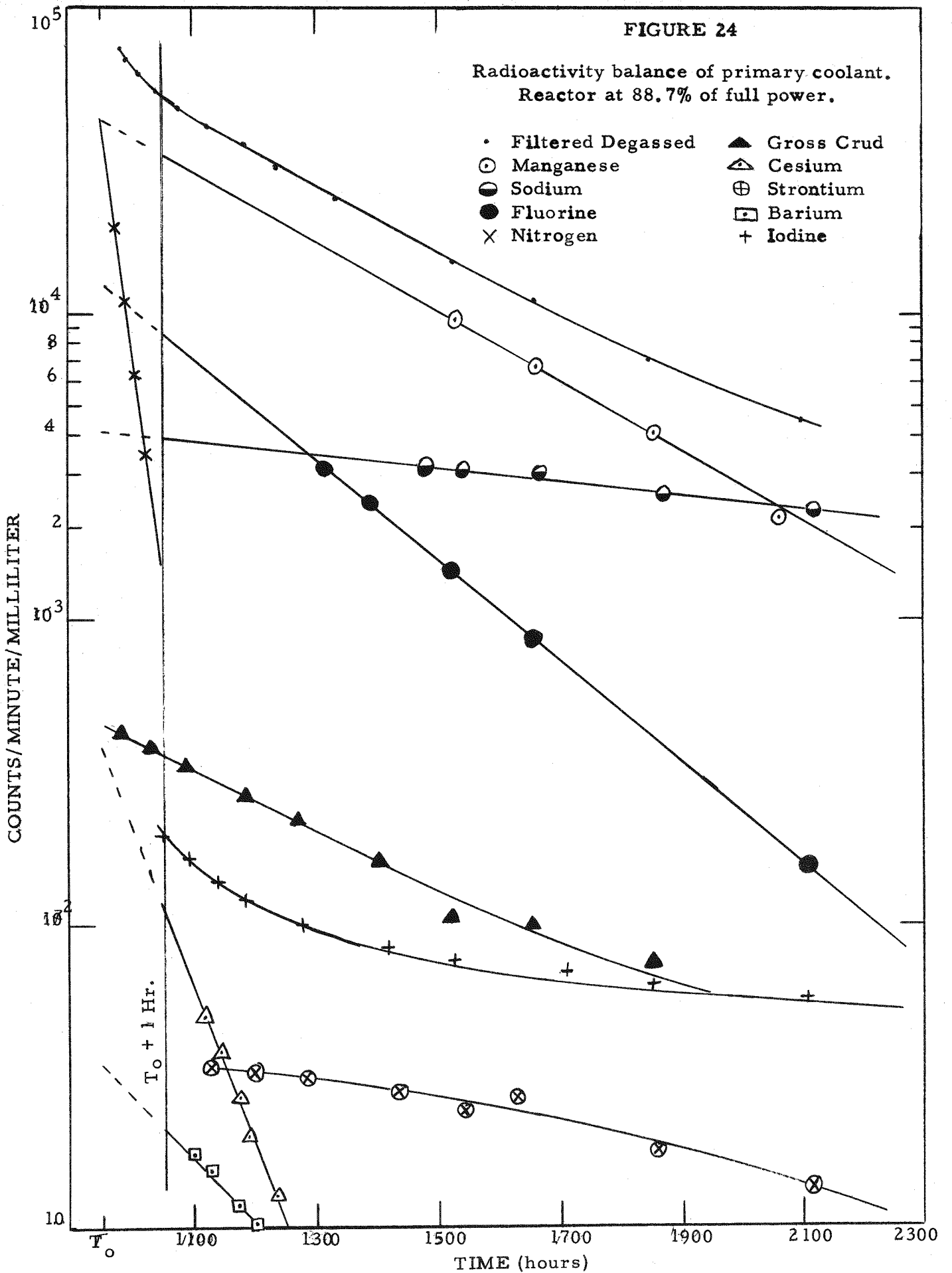
TABLE XI

RADIOACTIVITY BALANCE IN
PRIMARY COOLANT

Date: 16 March, 1962   Reactor Power: 9.7%

Time: 1424 hours   Time at Power: 550 minutes

Sample Measured	Concentration * (cpm/ml)	Percentage of Total
Gross filtered degassed	7.1×10^3	---
Mn^{56}	4.1×10^3	57.8
N^{13} (non-degassable)	2.4×10^2	3.4
F^{18}	1.9×10^3	26.2
Na^{24}	3.5×10^2	4.9
Cs^{138}	1.7×10^1	0.2
Gross I	3.2×10^1	0.5
		93.0
Gross Crud	2.5×10^2	

*Corrected to decay time of one hour after sampling.

TABLE XII

RADIOACTIVITY BALANCE IN
PRIMARY COOLANT

Date: 18 March, 1962

Reactor Power: 38.9%

Time: 1349 hours

Time at Power: 645 minutes

Sample Measured	Concentration * (cpm/ml)	Percentage of Total
Gross filtered degassed (total activity)	2.7×10^4	--
N^{13} (non-degassable)	3.4×10^2	1.3
F^{18}	4.1×10^3	14.9
Mn^{56}	2.1×10^4	77.6
Na^{24}	1.1×10^3	4.0
Gross I	7.4×10^1	0.3
Cs^{138}	3.1×10^1	0.1
Ba^{139}	1.0×10^1	< 0.1
Gross Sr.	1.4×10^1	< 0.1
		98.2

*Corrected to decay time of one hour after sampling.

TABLE XIII

RADIOACTIVITY BALANCE IN
PRIMARY COOLANT

Date: 24 March, 1962

Reactor Power: 53%

Time: 1415 hours

Time at Power: 497 minutes

Sample Measured	Concentration * , ** (cpm/ml)	Percentage of Total
Gross degassed *** (total activity)	4.2×10^4	--
N^{13} (non-degassable)	4.3×10^2	1.1
F^{18}	5.1×10^3	12.2
Mn^{56}	3.5×10^4	84.4
Na^{24}	2.1×10^3	5.0
Gross I	1.1×10^2	0.3
Cs^{138}	4.4×10^1	0.1
Gross Crud	2.3×10^2	0.6
		103.7

*Corrected to a decay time of one hour after sampling.

**Analyses performed at sea. Uncertainty in chemical yields due to severe
   vibrations of trailers and balance.

***This sample activity balance was performed on the basis of a gross de-
   gassed aliquot of primary coolant in place of the gross filtered degassed
   sample. With the addition of the crud contribution to the actual activity
   balance the samples are comparable.

TABLE XIV

RADIOACTIVITY BALANCE IN PRIMARY COOLANT

Date: 4 April, 1962   Reactor Power: 77.6%

Time: 1342   Time at Power: 777 minutes

Sample Measured	Concentration * (cpm/ml)	Percentage of Total
Gross filtered degassed	3.7×10^4	---
Mn^{56}	2.4×10^4	65.2
F^{18}	5.2×10^3	14.2
N^{13} (non-degassable)	1.0×10^3	2.7
Na^{24}	2.8×10^3	7.5
Gross I	1.4×10^2	0.4
Cs^{138}	6.5×10^1	0.2
Ba^{139}	2.6×10^1	---
Ba^{140}	1.0×10^0	---
		90.2
Gross Crud	2.1×10^2	

*Corrected to a decay time of one hour after sampling.

TABLE XV

RADIOACTIVITY BALANCE IN PRIMARY COOLANT

Date: 5 April, 1962

Reactor Power: 88.7%

Time: 0932 hours

Time at Power: 512 minutes

Sample Measured	Concentration * (cpm/ml)	Percentage of Total
Gross filtered degassed	5.0×10^4	---
Mn^{56}	3.5×10^4	68.8
Na^{24}	3.9×10^3	7.7
F^{18}	8.8×10^3	17.4
N^{13} (non-degassable)	1.2×10^3	2.5
Gross I	1.9×10^2	0.4
Cs^{138}	1.2×10^2	0.2
Ba^{139}	2.1×10^1	---
		97.0
Gross Crud	3.6×10^2	

*Corrected to a decay time of one hour after sampling.

D. Waste Tank and Demineralizer Effluent Studies

At selected intervals, or at the request of the shift Health Physicist, measurements were made of the radioactivity in the waste tank and the demineralizer effluent. Traces of radioactivity were observed in waste tank samples as the reactor approached full power. The observed concentration of radioactivity was in every case below the maximum permissible concentration for discharge of the contained liquids to the environment. The total radioactivity concentrations were too small ($< 10^{-7}$ μc/ml) to permit accurate identification and measurement of specific radionuclides. However, it is believed that Na^{24} was the principal radioactive constituent.

A comparison of a gamma spectrum obtained with a one-liter sample of primary coolant obtained at the demineralizer effluent to that of a one-liter sample obtained at the demineralizer influent is shown in Figure 25. The principal photopeak in the effluent sample was due to the 1.28-Mev photopeak of Ar^{41}. It may be seen, from Figure 25, that the Ar^{41} concentration was identical in the influent and effluent samples. Since the demineralizer is not effective in removal of noble gases from the primary coolant, this behavior is normal.
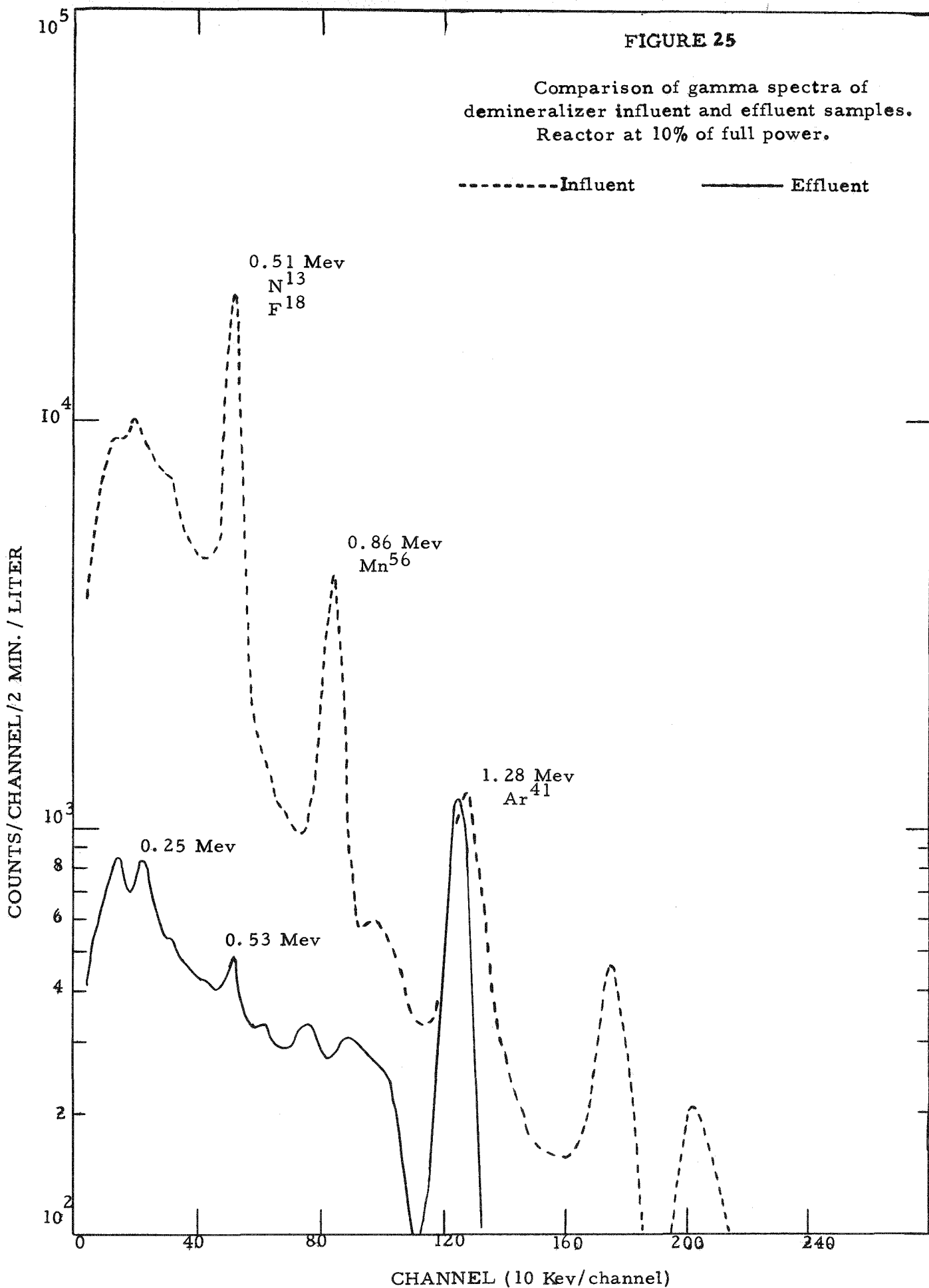
In addition to the photopeak due to the 1.28-Mev photon of Ar^{41}, four other prominent photopeaks were observed in the influent sample. Three of these peaks, at 0.86 Mev, 1.81 Mev, and 2.13 Mev, correspond to gamma rays emitted by Mn^{56}. The photopeak at 0.51 Mev is attributed to N^{13} and F^{18}. Comparison of the spectra in Figure 25 shows that the concentrations of these latter three nuclides were reduced at least a factor of ten by the demineralizer.

Several small photopeaks corresponding to photon energies less than 1.28 Mev were also present in the spectrum obtained for the effluent sample. The photopeak at 0.53 Mev is believed to be due to Xe^{135m}. Although it is difficult to identify the specific radionuclides which gave rise to the other photopeaks, evidence exists that they may be due to the noble gas fission products, Kr^{85m}, Kr^{88}, and Xe^{135}. The effluent sample with which the spectrum in Figure 25 was obtained was subsequently degassed and then reanalyzed. No significant gamma radioactivity was found in the degassed effluent sample. This behavior was typical of the gamma radioactivity in effluent samples. Therefore, it is concluded that the efficiency of the demineralizer for removal of cationic or anionic fission products from the primary coolant was greater than 90%.

FIGURE 25

Comparison of gamma spectra of
demineralizer influent and effluent samples.
Reactor at 10% of full power.

--------Influent ———— Effluent

0.51 Mev
N^{13}
F^{18}

0.86 Mev
Mn^{56}

1.28 Mev
Ar^{41}

0.25 Mev

0.53 Mev

COUNTS/CHANNEL/2 MIN. / LITER

CHANNEL (10 Kev/channel)

- 58 -

IV. DISCUSSION

IV. DISCUSSION

The radiochemistry investigations performed during the start-up and initial phases of operation of the N. S. Savannah reactor plant have provided data for the radioactivity content of the primary coolant and other portions of the reactor plant (primary and secondary systems).   These data may be applied to the evaluation of performance of specific components and systems of the reactor and may be used as a basis for evaluating future reactor plant operations.

Ultra-sensitive analyses for radioactive species in samples of the primary coolant, in fluids from secondary coolant and auxiliary systems, and in sources of make-up water at the Camden, N. J., location showed conclusively that no significant radioactivity was present in any of these fluids prior to initial criticality of the reactor.   On the basis of these data (Table III), it has been concluded that the elaborate precautions taken during core loading operations to minimize inadvertent introduction of foreign objects into the reactor were also effective in excluding radioactive fallout from the system.   Therefore, no significant contamination of the primary system resulted from radioactive debris introduced by the 1961 U. S. S. R. atmospheric nuclear tests.

Prior to its installation aboard the vessel, the reactor core had been subjected to criticality experiments which might have resulted in production of small quantities of radioactivity in the core or in radioactive contamination of the core surfaces.   Damage to a fuel element during core loading operations could have resulted in the release of radioactivities to the fresh coolant.   Furthermore, radioactive contaminants on the core surface may have been leached off by the coolant. The net results of the analyses of pre-criticality samples showed that no significant radioactivity had been introduced into the primary coolant from any of these sources and that the procedures established for pre-criticality operations had not resulted in any damage to the core detectable by the analytical methods used.   Since the plant coolants were shown to be free from significant concentrations of radioactivity prior to criticality, any radioactivity observed subsequent to criticality can be attributed to reactor operation.

A measurable concentration (0.12 μg/l) of uranium was present in the primary coolant at initial criticality. The source of this uranium has not been established, but it is believed to be a chemical impurity in the make-up water. Periodic measurements showed that the values of the concentration decreased with continued reactor operations. This decrease is believed to be due to the removal of the uranium by the demineralizer or by deposition on primary system surfaces. These trace concentrations of uranium in the coolant established a minimum value for the concentrations of fission products in the coolant. This subject is discussed below in conjunction with other aspects of the fission product studies. It is recommended that periodic measurements of the uranium concentration in the primary coolant be made as a portion of the continuing surveillance of the reactor plant.

Data obtained during daily surveillance of the primary coolant showed that no unusual or abnormal change in the concentration of gross gamma radioactivity occurred at any time throughout these studies. The gradual increase in this concentration after normalization to 100% of full power as the reactor operated at increasing power levels was not unexpected. As the reactor continued to operate near its rated maximum power output, the mean value of the normalized concentration of the gross 15-minute degassed radioactivity was $(1.2 \pm 0.2) \times 10^{-1}$ μc/ml. This value is established as the base operational level for subsequent plant operation.

It is anticipated that the concentrations of gross 15-minute degassed radioactivity may increase gradually with increased reactor operation. Such an increase might be due to such factors as the normal gradual increase in the contribution of longer lived radionuclides to the total gamma radioactivity or an increase in the concentrations of soluble or particulate corrosion products. However, a precipitous increase in the concentration of the gross gamma radioactivity is usually an indication of a potential hazard due to a faulty core component or a change in the chemical composition of characteristics of the primary coolant. If any sudden increase should occur, immediate steps should be taken to define quantitatively the contributing species.

The results of the routine measurements of the concentration of gross one-hour iodine radioactivity in the primary coolant showed that no precipitous increase in fission product concentrations had occurred either during steady-state operations or as a result of reactor power transients. The concentration of gross one-hour iodine radioactivity remained below the maximum permissible concentration for discharge to the sea[8]. The mean value of the normalized concentrations observed at reactor power levels in excess of 10% of full power was $(2.0\pm0.3)\times10^{-4}$ $\mu c/ml$. This value, which is defined as the base concentration for subsequent reactor operations, is consistent with values for the concentrations of gross one-hour iodine radioactivity observed in other pressurized water reactor coolants in which the integrity of the fuel cladding has been maintained.

It is possible that the concentration of gross one-hour iodine radioactivity may increase slowly over the first several thousand equivalent full-power hours, and then remain essentially constant. This possible future increase could result from the conversion of U^{238} on the core surface to Pu^{239}. However, any sudden increase in the concentration of the gross one-hour iodine radioactivity would be a specific indicator of a significant influx of fission products (e.g., through a fuel element cladding failure) and warrants immediate investigation.

If such an increase occurs concurrently with an increase in the gross radioactivity concentration, fission products will have been established as the principal contributors to the increase. Further detailed identification of the particular fission products and determinations of their concentrations can then be made either by gamma spectrum or radiochemistry methods. However, if no increase in the gross one-hour iodine concentration occurs in conjunction with a serious increase in gross gamma radioactivity, fission products may, in general, be excluded as principal contributors to the observed increase. In such an eventuality, attention may then be focused on activation products or activated corrosion products to determine the events which gave rise to the increased radioactivity.

The absence of sudden increases in the gross one-hour iodine concentration during a sustained period of reactor operations does not necessarily preclude the

existence of a slightly defective fuel element in the core. Frequently such increases occur at plant start-up or shutdown, or as a result of power transients. Throughout this program, measurements were made of the gross one-hour iodine radioactivity immediately following reactor power transients. In no instance was any evidence obtained for a significant fuel cladding defect. It is recommended that this indicator of fuel cladding failure be used routinely aboard the vessel, especially during periods of power transients, to continue surveillance of fuel cladding integrity.

In addition to the daily surveillance operations, extensive studies were performed to obtain detailed characterization of the important radioactive constituents of the primary coolant. A total of 22 radionuclides were measured quantitatively during these studies. Four of the radionuclides were activation products, two of which (F^{18} and N^{13}) are intrinsic to any water-cooled reactor. Due to the short half-lives of these two species, their concentrations are proportional to reactor power. The mean value of the concentration of F^{18} observed at the various reactor power levels, after correction for decay and normalization to 100% of full power, was $(1.6\pm0.1)\times10^{-2}$ $\mu c/ml$. Since this value exhibits a standard deviation of less than 10%, it is concluded that the F^{18} concentration exhibited normal behavior during the period in which these measurements were performed.

Analysis for N^{13} in the primary coolant is somewhat more complex, since it may be present both as a free gas (N_2) and in a chemically combined form, such as ammonium ion $(NH_4)^+$. Measurements of the chemically combined, or non-degassable, nitrogen in samples of the primary coolant resulted in a mean value for the normalized N^{13} concentration of $(9.5\pm0.9)\times10^{-2}$ $\mu c/ml$. Attempts to measure quantitatively the concentration of the degassable N^{13} in these samples were not satisfactory. However, the apparent values, which were very small, may not be representative of the actual concentration of volatile N^{13} in the primary coolant because of potential losses resulting from the depressurization of the coolant upstream of the coolant sampling facility. Direct measurement of the total N^{13} could not be made because the shipboard gas sampling system was inoperative during the period in which these measurements were desired. Therefore, the data for the N^{13} represent lower limits for its concentration in the primary system.

An apparent preferential distribution of N^{13} into the chemically-combined state in the primary coolant of the N. S. _Savannah_ might be postulated as follows: Since the pH of the coolant is approximately 6.5, an excess of hydrogen ions exists. In addition, a significant concentration of hydrogen radicals resulting from radiolysis of the coolant may be present. These two species may react with the recoil nitrogen ions produced as a result of $O^{16}(p, \alpha)N^{13}$ to form ammonia or ammonium ions. This chemical reaction may compete favorably with the reactions in which the nitrogen ions recombine to form elemental nitrogen.

The other two significant activation products, Na^{24} and Ar^{41}, result from activation of sodium and argon impurities in the primary coolant. Since the concentrations of these impurities are not necessarily constant, a direct proportionality between reactor power and their concentrations may not be observed. With reference to Table V, it is seen that the Na^{24} concentration exhibited an increasing trend with continued reactor operations. The observed concentrations were, however, consistent with concentrations observed in similar reactors. Since sea-water is used as a source of make-up water for this reactor, the concentrations of Na^{24} observed are judged to be normal.

Argon is normally present in trace concentrations in the primary coolant, since it has a finite solubility in water. Therefore, its concentration, after normalization to maximum reactor power, is expected to be constant under normal operating conditions. The observed values of the concentration of Ar^{41} in the primary coolant samples were very small, and were subject to the same uncertainties as the data for N^{13}. Attempts were made to estimate the Ar^{41} losses due to the depressurization in the sampling line by comparing data for degassed and non-degassed coolant samples. The results of these comparisons were inconclusive. It is believed, therefore, that these data are not truly representative of the actual concentrations of Ar^{41} in the primary coolant of the N. S. _Savannah_.

In addition to its normal content in the primary coolant, argon can be introduced into the primary system by air leakage. Since an increased concentration of free oxygen in the system can adversely affect corrosion rates and crud formation and deposition, the existence and application of a rapid indicator for significant

increases in air concentrations in the system is desirable. The Ar^{41} concentration in the coolant provides such an indicator. However, the valid measurement of Ar^{41} requires acquisition of a truly representative sample of the primary system. The availability of an operable pressurized sampling facility for this purpose is, therefore, deemed necessary for complete monitoring of this important plant parameter.

The very short-lived (9.7-second) isotope N^{16}, is also a normal activation product in pressurized water reactors. Analysis for this species as a part of these studies was deemed desirable. However, the transport time of the coolant from the reactor core to the coolant sampling facility was close to 80 half-lives of this species. The resulting extensive in-transit decay precluded its measurement.

During the course of this work, analyses were performed for eight activated corrosion products. The concentration of Mn^{56} observed at each power level represented the principal fraction of the gross gamma radioactivity. It was observed to be the most important radioactive constituent in both the aqueous and separated crud fractions of primary coolant. No other activated corrosion product was observed in significant concentrations at power levels below 38.9% of full power. At this power, Cu^{64} was also present in the crud fraction in measurable concentrations. Analyses for other nuclides in this category showed that their concentrations in the coolant and crud were either insignificant or extremely small. The values for the long-lived species (Table VI) were derived from sensitivity measurements and are subject to rather large statistical errors due to the low counting rates. In most of these analyses, precisions of only ± 30% were achieved.

Tungsten-187, a radioactive constituent common to the primary systems of most pressurized water reactors, was not observed in either the aqueous or particulate fraction of the coolant at any power level. The absence of significant quantities of W^{187} is consistent with the results of a chemical analysis of particulate samples obtained prior to initial criticality[10]. No evidence for the presence of tungsten was obtained in these analyses.

The distribution of the activated corrosion products between the aqueous and particulate phases of the primary coolant is shown in Table VI. The manganese isotopes were observed to be associated primarily with the aqueous fraction. At each

power level at which studies were performed, 90% or more of the Mn^{54} and Mn^{56} was found in the aqueous fraction. At power levels less than 20% of full power, 90% to 95% of the observed manganese isotopes were present in the aqueous fraction. The Fe^{59} was found to be associated preferentially with the crud fraction, as were $Co^{58,60}$, and Cr^{51}. The chromium and cobalt isotopes exhibited a pronounced affinity for the particulate fraction in the sample taken when the reactor was operating at 100% of full power. Approximately 80% of the Fe^{59} was observed to be in the particulate fraction.

The 12.8-day Cu^{64}, which first was detected in the crud fraction at 38.9% of full power, was not generally sought in the aqueous fraction. An analysis for this nuclide, performed on the aqueous fraction obtained at 53.8% of full power, showed that no significant concentration was present at that time.

The concentrations of the activated corrosion products in the primary coolant exhibited an increasing trend with increasing reactor power. These increases in concentrations generally proceeded at a rate faster than can be attributed to increased power or equivalent full-power hours of operation. This larger increase is not unexpected, since corrosion products were continually introduced into the coolant during this period. The concentrations were generally very small. The concentrations of Mn^{56} were somewhat larger than those which have been observed in similar reactors. However, Mn^{56} does not appear to constitute any potential hazard.

No abnormal variations in activated corrosion product concentrations were observed during this program. No evidence was obtained for radioactivity transients such as those associated with sudden releases of corrosion products or particulates to the coolant (crud bursts), or for abnormal changes in primary system corrosion.

During the course of this work, analyses were performed on primary coolant samples for 10 fission products. The concentrations of the iodine isotopes, $I^{131, 133, 134, 135}$, increased slightly with reactor power as is shown in Table VII. The increases were consistent with, or somewhat less than, the increases in reactor power. The variation in the concentration of 32-minute Cs^{138} in the primary coolant was proportional to the reactor power within experimental error.

The concentrations of the short-lived strontium isotopes, 9.7-hour Sr^{91} and 2.7-hour Sr^{92}, in the filtered samples of primary coolant (Table IX) exhibited little correlation with reactor power. The concentration of Ba^{139} in the aqueous fraction of the primary coolant also exhibited fluctuations not readily correlated with reactor power. The fluctuations for these three species are believed to be due in part to their low counting rates in the separated fractions. Also, uncertainties were introduced into the chemical yields for those fractions separated from samples taken at the higher reactor power levels by difficulties encountered in using the analytical balance at sea. However, the data for Ba^{140} concentrations appear to be consistent with changes in reactor power.

No significant concentrations of barium or strontium isotopes were observed in the crud fractions. It has been shown previously[11] that the distribution of barium and strontium isotopes between the aqueous and particulate fractions of the primary coolant of a pressurized water reactor is largely dependent upon the pH of the primary coolant. As the value of the coolant pH increases above 8, the barium and strontium isotopes tend to be associated increasingly with the particulate fraction of the coolant. Since the primary coolant of the N. S. Savannah was maintained between pH-6 and pH-7 at power levels above 18% of full power, the preferential distribution of barium and strontium into the aqueous phase is not unexpected.

An estimate was made of the concentrations of the above fission products which would result from fission of the observed trace concentrations of uranium in the primary coolant. A maximum contribution of the order of 1% of these concentrations is attributed to this source. Since this contribution is negligible, and since the data for gross one-hour iodine radioactivity indicated that there was no significant defect in a fuel element cladding, the fission products observed in the primary coolant were assumed to have resulted from fission product recoils from natural uranium contamination on reactor core surfaces. On the basis of this assumption, the experimentally determined values of the concentrations of our pre-selected fission products have been used to estimate the uranium contamination on the core surfaces.

The four fission products, I^{133}, I^{134}, Cs^{138}, and Ba^{139}, were selected on the basis of the criteria delineated in Section I. Their concentrations were measured at nearly steady-state conditions, as reactor operations at several reactor power levels permitted. The experimental values were corrected to the steady-stated concentrations and were normalized to the maximum rated reactor power. The surface density of uranium on the core heat transfer surfaces, assuming natural uranium, was computed using equation (11), Appendix I. The data are presented in Table XVI. The mean value of the surface density of natural uranium contaminant on the heat transfer surfaces of the core was $(5.6\pm1.1)\times10^{-2}$ $\mu g/cm^2$. If this contamination were due entirely to fully-enriched (93% U^{235}) uranium, the apparent surface density would be 4.3×10^{-4} $\mu g/cm^2$. This level of uranium contamination does not appear to represent any potential difficulty in plant operations.

In the radioactivity balance studies, measurements of the concentrations of the gamma radioactive species in the chemically separated fractions agreed, within experimental error, with the total gamma radioactivity concentrations in the sample. These results confirm that all significant radionuclides in these samples have been identified and measured quantitatively. It is recommended that similar studies be performed periodically during subsequent reactor operations to confirm the results of the more routine surveillance of the reactor plant.

TABLE XVI

APPARENT URANIUM CONTAMINATION OF
REACTOR CORE HEAT TRANSFER SURFACES

Nuclide	Fission Yield (%)	Reactor Power (% of Full Power)	Time at Power (min.)	Surface Density Uranium ($\mu g/cm^2$)
I^{134}	7.8	9.7	300	6.0×10^{-2}
		18.0	550	5.6×10^{-2}
		38.9	645	6.0×10^{-2}
		53.0	497	5.4×10^{-2}
		77.6	777	5.4×10^{-2}
		88.7	512	6.4×10^{-2}
I^{133}	6.9	9.7	300	5.1×10^{-2}
		38.9	645	5.5×10^{-2}
		53.0	497	3.0×10^{-2}
		77.6	777	3.4×10^{-2}
		88.7	512	8.2×10^{-2}
Cs^{138}	5.6	9.7	320	6.3×10^{-2}
		18.0	550	6.3×10^{-2}
		38.9	645	6.7×10^{-2}
		53.0	497	6.6×10^{-2}
		77.6	777	7.5×10^{-2}
		88.7	512	7.5×10^{-2}
Ba^{139}	6.6	18.0	550	4.6×10^{-2}
		38.9	645	3.7×10^{-2}
		53.0	497	5.1×10^{-2}
		77.6	777	5.1×10^{-2}
		88.7	512	3.6×10^{-2}

Overall average $(5.6 \pm 1.1) \times 10^{-2}$

V. SUMMARY

V. SUMMARY

This program has demonstrated that the radioactivity content of the primary system of the N. S. Savannah reactor plant was small and normal during the period of initial criticality and start-up, and during the sea trials and acceptance tests. The principal radioactive constituents (Mn^{56}, Ar^{41}, N^{13} and F^{18}) are either intrinsic to the primary system of the pressurized water reactor or are normally found in the coolant in concentrations comparable to those observed in this program. The Mn^{56} concentrations observed at the various reactor power levels were slightly higher, relative to those for the other nuclides, than those observed in similar reactor plants. This slightly increased concentration is attributable to the fact that the coolant of this reactor was generally maintained between pH6 and pH7, whereas the primary coolants of the other plants were maintained at somewhat higher pH values.

Data for fission product concentrations in the primary coolant indicate that their only significant source is uranium contamination of the reactor core surfaces. The observed concentrations do not represent any significant hazard or potential difficulty in plant operation. The small value of 5.6×10^{-2} $\mu g/cm^2$ for the surface density of uranium indicates that no significant contamination of these surfaces occurred during core fabrication. No significant defect in a fuel element cladding was detected during the period in which these measurements were performed.

The efficiency of the demineralizer for removal of anionic and cationic radionuclides from the primary coolant was shown to exceed 90%. Volatile radionuclides were the only radioactive constituents found in the demineralizer effluent.

Data obtained for the concentrations of gross radioactivity in the waste tanks were maintained below the maximum permissible concentrations for discharge to the environment.

On the basis of these radiochemistry studies, it may be concluded that the N. S. Savannah primary coolant system exhibited a normal content and distribution of radionuclides and that those plant components for which radioactivity indicators or monitors were observed exhibited safe and normal operation behavior.

APPENDIX I

APPENDIX I

CALCULATION OF THE URANIUM CONTAMINATION
ON REACTOR CORE SURFACES

The quantity, $N(t)$, of a radioactive fission product in the primary system of a pressurized water reactor at a particular time, t, is a function of its rate of introduction into the coolant, its rate of decay, and the rate at which it is removed by the purification system. Therefore, the rate equation describing the build-up of the species in the coolant is given by equation (1).

$$\frac{dN(t)}{dt} = R - \lambda N(t) - \frac{F_b N(t)}{V_p} \tag{1}$$

where
$N(t)$ = the number of atoms of the fission product in the primary coolant;

R = the rate of introduction into the coolant, sec-1;

λ = the decay constant, sec-1;

F_b = the bypass flow rate at operating temperature through the decontamination system, $cm^3/sec.$;

V_p = the volume of primary coolant, at operating temperature, cm^3; and

F_b/V = β, the plant purification constant, sec-1.

Integration of equation (1) yields:

$$N(t) = \frac{R}{(\lambda+\beta)} \left(1 - e^{-(\lambda+\beta)t}\right). \tag{2}$$

At steady state conditions (i.e., $t = \infty$),

$$N^\infty = \frac{R}{(\lambda+\beta)} , \tag{3}$$

and the disintegration rate of the fission product, in terms of atoms disintegrating per second, at steady state is

$$D^{\infty} = R \frac{\lambda}{(\lambda + \beta)} = D^{\infty}_v V_p , \qquad (4)$$

where D^{∞}_v is the steady-state concentration of the fission product in the primary coolant, (dps/cm^3).

Therefore,

$$R = \frac{(\lambda + \beta)}{\lambda} D^{\infty}_v V_p . \qquad (5)$$

If the source of the fission product is uranium contamination on core surfaces, the rate of introduction is the rate at which the fission product recoils are released to the coolant. This rate is a function of the quantity of effective uranium, the mean neutron flux to which the uranium is exposed, and the fission yield of the particular nuclide. The effective uranium is that uranium contained within a distance from the surface equal to one recoil range.

If it is assumed that one-fourth of the fission recoils reach the coolant, the recoil rate is given by:

$$R = \frac{\phi_m \, n_{235} \, Y\sigma_f}{4} , \qquad (6)$$

where ϕ_m = the mean neutron flux ($= 7.15 \times 10^2$ n/cm^2-sec at 100% of full power);

n_{235} = the number of effective atoms of U^{235};

Y = the fractional fission yield of the radionuclide; and

σ_f = the thermal neutron fission cross-section of U^{235} ($5.82 \times 10^{-22} cm^2$).

Substitution of equation (6) into (5) yields:

$$n_{235} = \frac{4 \, V \, D^{\infty}_v}{\phi_m \sigma_f \, Y} \frac{(\lambda + \beta)}{\lambda} . \qquad (7)$$

Equation (7) may be expressed in terms of natural uranium by:

$$n_U = \frac{5.6 \times 10^2 \ V \ D_v^\infty}{\phi_m \ \sigma_f \ Y} \qquad \frac{(\lambda + \beta)}{\lambda} \ . \tag{8}$$

The quantity of effective uranium may be expressed in terms of micrograms per unit area of core surface, or

$$m_U = 3.96 \times 10^{-16} \qquad \frac{(n_U)}{A} \ . \tag{9}$$

Therefore,

$$m_U = \frac{2.2 \times 10^{-13} V \ D_v^\infty}{\phi_m \ \sigma_f \ Y \ A} \qquad \frac{(\lambda + \beta)}{\lambda} \ . \tag{10}$$

If the measured values of D_v^∞ are normalized to 100% of full reactor power, and if the corresponding values of ϕ_m and of σ_f are substituted in equation (10), the resulting relationship $m_U = 5.35 \times 10^{-5} \ \frac{(V \ D_v^\infty)}{A \ Y} \quad \frac{(\lambda + \beta)}{\lambda}$ (11) may be used to calculate the quantity of natural uranium per unit area of core within one recoil range of the surface. This value is termed the uranium surface contamination.

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