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IDENTIFICATION, SOURCE AND CONTROL

D. W. Moeller*

*Radiological Health Program, Public Health Service, U. S. Department of Health, Education, and Welfare, assigned to the Oak Ridge National Laboratory

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ACKNOWLEDGMENTS

Revise the acknowedgment page, iii, to read:

This report is based upon a thesis submitted to North Carolina State College in partial fulfillment of the requirements for the doctoral degree. The report describes research carried out in the Analytical Chemistry Division at the Oak Ridge National Laboratory.

The research was supported by the Oak Ridge Graduate Fellowship Program of the Oak Ridge Institute of Nuclear Studies and was directed by a committee appointed by Dean D. B. Anderson of North Carolina State College Graduate School which was composed of Dr. Newton Underwood and Mr. Joseph Lynn of the Department of Physics at North Carolina State College. James G. Terrill, Radiological Health Program, Division of Sanitary Engineering Services, U. S. Public Health Service, and Dr. Myron T. Kelley of the Analytical Chemistry Division and Mr. Roy J. Morton of the Health Physics Division of the Oak Ridge National Laboratory.

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CHAPTER I

STATEMENT OF THE PROBLEM

Reactor cooling water systems, currently employed, are basically of two types: (1) single pass and (2) recirculating. Typical of reactors employing single pass systems are those at the Hanford Works of the Atomic Energy Commission (Conley, 1954), the original Raleigh Research Reactor (Beck, et al., 1950) and the proposed Test Tube Research Reactor (Busey and Hammond, 1955). Typical of reactors utilizing recirculating cooling water systems are the Low Intensity Test Reactor and the new Oak Ridge Research Reactor at the Oak Ridge National Laboratory, the Materials Testing Reactor at the National Reactor Testing Station, the pressurized Water Reactor (AEC, 1954) at Shippingport, Pennsylvania, and the S2W nuclear propulsion plant of the USS Nautilus (AEC, 1956).

Cooling water utilized in both type systems becomes radioactive. The resulting radioactive products are thought to originate from: (a) activation of impurities (both dissolved solids and gases) present in the water, (b) activation of the water itself, (c) corrosion and scale formation and (d) fuel and fission products escaping into the water from the reactor fuel elements.

The purpose of this work was to study, in detail, a reactor cooling system of the recirculating type. Principal objectives of the study were identification of the radioactive nuclides present in the water at various stages in the system, determination of

the source of these nuclides and measurement of the efficiency of ion exchange beds in their removal.

Identification of the radionuclides in the cooling water would furnish information as to their source. Establishment of their source could then lead to effective control, thus reducing accessibility problems and contamination hazards. Monitoring of the radioactive material in the cooling water would furnish information warning of unusual circumstances within the reactor system, such as rupture of a fuel element, failure of an experimental facility (Heath 1956) or excessive corrosion. Early detection of such conditions is essential to safe reactor operations. These data and data related to the efficiency of the ion exchange beds in the decontamination of cooling water are of importance to the nuclear engineer, as well as public health personnel who are concerned with unnecessary and/or accidental release of radioactive materials to the environment.

Supplemental objectives of the study included evaluation of neutron activation techniques as applied to water and evaluation of gamma scintillation spectrometric techniques in the qualitative and semiquantitative determination of radionuclides in water. Although primarily directed here toward the cooling water problem, the results of these evaluations, too, should be of interest to public health personnel who more and more frequently are called upon to determine small concentrations of impurities in drinking water and to identify and measure radioactive materials in streams and lakes.

CHAPTER II

BASIC TYPES OF COOLING SYSTEMS

A. Single Pass

In the single pass system the cooling water passes through the reactor only once and then is discharged. Where the source is the local city supply, the water may be utilized with no special additional treatment. Where the source is a lake or river, the water may receive extensive chemical and/or ion exchange treatment prior to use. In all such systems, a retention basin is needed to permit decay of radioactive material in the discharged cooling water prior to its release to the surrounding environment. The single pass system is limited to reactors located in areas having unusual sources of rather pure water and providing ample dilution for the effluent.

Figure 1 is a schematic representation of the single pass system (Woodruff, et al., 1953).

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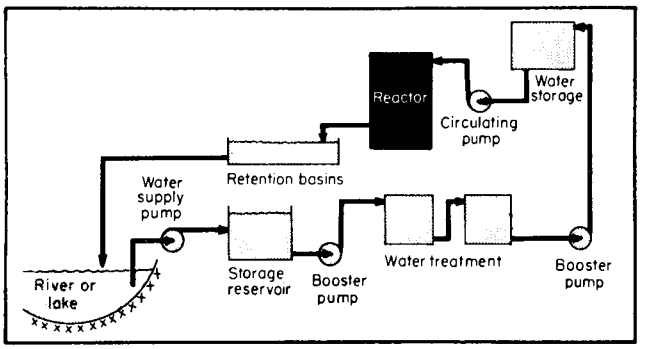

Fig. 1. Schematic Flowsheet: Single Pass Cooling Water System.

B. Recirculating

In the recirculating type the cooling system is originally filled with high purity water such as that obtained by distillation or demineralization. Deaeration of make-up water may also be provided. Once the system is filled, the water is recycled within the closed unit, its quality being maintained by continuous operation of an ion exchange purification system through which a small portion of the water is bypassed. Radioactive material removed from the water is retained on the ion exchange resins until removed by regeneration. Regenerating solutions should be stored to prevent unnecessary release of radioactive material to the environment. Recirculating systems are satisfactory for most reactor types.

Figure 2 is a schematic presentation of the recirculating system (Woodruff, et al., 1953).

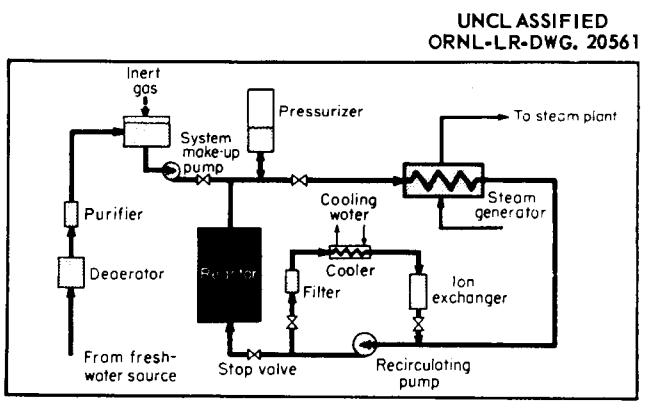

Fig. 2. Schematic Flowsheet: Recirculating Cooling Water System.

Various modifications and combinations of the two basic systems also are used. For example, systems exist in which the

water is recirculated, its quality being maintained by daily release of a small amount to waste. The volume of contaminated water discharged is replaced by high purity make-up water.

CHAPTER III

PREVIOUS RELATED STUDIES

Studies have been made relating to the radioactive products in water cooling systems of both the single pass and recirculating types. Sivetz (1950) discussed the sources of radioactive nuclides in water and certain radioactive nuclides which would be induced through neutron irradiation of impurities common to natural waters. His work showed 7.4 s N^{16}, 29 s O^{19}, 5.1 m Cu^{66}, 9.5 m Mg^{27} and 2.3 m Al^{28} to be important short-lived activities. Both single pass and recirculating systems were discussed and the author concluded that the more logical design for future reactors was the closed cycle type.

The main source of laboratory data on single pass cooling systems is that which has been obtained on the Columbia River. This river receives cooling water from the reactors at the Atomic Energy Commission's Hanford Works, which is operated by the General Electric Company. Conley (1954), Foster and Rostenbach (1954) and Robeck, et al. (1954) have published data concerning radioactivities in the Columbia River. The predominant nuclides identified in the water, as determined by chemical separation and beta absorption and decay studies, were 15.0 h Na^{24}, 2.58 h Mn^{56}, 14.5 d P^{32}, 27 d Cr^{51}, 12.8 h Cu^{64}, 2.6 h Si^{31} and 26.7 h As^{76}. Certain of the fission products, namely, 54 d Sr^{89}, 28 y Sr^{90} and 12.8 d Ba^{140}, were also reported as being observed in lesser concentrations.

This author (Moeller, 1956) made a theoretical investigation of the induced radioactive nuclides to be expected in the effluent cooling water from the Hanford reactors and found a close correlation of the theoretical data with published laboratory data. These calculations revealed that, as far as induced radioactive nuclides were concerned, the critical cationic elements in water were sodium, arsenic, manganese and copper. The critical anions were chloride, sulfate, phosphate and chromate. Of the dissolved gases in water, argon appeared to be the most significant. The studies further showed that, where the source of cooling water was the local city drinking supply, the resulting radioactivity in effluent cooling water would be influenced by many factors. Such factors include the coagulants employed at the local city water treatment plant; chemicals added to the water for purposes of controlling disease, tastes and odors, corrosion and scale formation, as well as dental decay; and chemicals present in the water due to seasonal variations in the raw water supply.

Similar calculations (Beck, et al., 1950) were made for use in the design of the single pass cooling system for the Raleigh Research Reactor. However, laboratory data on the radioactive nuclides present in the effluent cooling water from this reactor have not been published.

The major work on determination of radioactive nuclides in recirculating cooling water systems is that which was done corollary to the development of a method for the rapid detection of fuel

cladding failures in the Materials Testing Reactor (Heath, 1956). Initial studies were made at a time when no known fuel rupture of detectable proportions had occurred in the reactor. The reactor fuel was aluminum clad. Identification of the radioactive nuclides was based upon gamma spectral and decay studies coupled with chemical and ion exchange separation techniques. The major activities found in the water up to a few minutes after removal from the flux region were 7.4 s N^{16}, 2.3 m Al^{28}, 2.58 h Mn^{56} and 15.0 h Na^{24}. Following the decay of the sodium activity, the remaining radioactive nuclides were determined to be 5.2 y Co^{60}, 245 d Zn^{65}, 270 d Ag^{110}, 9.5 m Mg^{27}, 1.87 h F^{18}, 12.8 h Cu^{64}, 54 h Cd^{115}, 27 d Cr^{51}, 2.56 h Ni^{65}, 45 d Fe^{59} and 71 d Co^{58}.

The comprehensive work of Heath (1956) included studies of stainless and carbon steel experimental loops as well as the aluminum system discussed in the previous paragraph.

Similar work on the identification of radioactive nuclides in cooling water has been reported by Lyon and Reynolds (1955 a). Here again, gamma spectrometry, decay studies and radiochemical methods (Reynolds, 1955) were utilized in the identification of the active nuclides. The principal short-lived activities found were N^{16}, Al^{28} and Mg^{27}. Longer-lived materials included Np^{239}, Mn^{56}, Na^{24}, Cu^{64} and Ag^{110} (Lyon and Reynolds, 1955 b). The water came in contact with aluminum, stainless steel, cast iron and carbon steel.

CHAPTER IV

RECIRCULATING SYSTEM OF THE LOW INTENSITY TEST REACTOR (LITR)

Because of its availability to the author, the cooling system of the Low Intensity Test Reactor (LITR) of the Oak Ridge National Laboratory (ORNL) was chosen for these studies.

The LITR was originally built to serve as the mock-up of the Materials Testing Reactor. However, after being used for mechanical and hydraulic studies and later for criticality measurements, the mock-up was subsequently converted into a general experimental facility and has operated as such since March, 1951.

The LITR is a water moderated and cooled, beryllium reflected, heterogeneous, enriched uranium reactor. The fuel elements are of aluminum-uranium alloy with aluminum cladding. The active lattice measures approximately 9 inches x 24 inches x 24 inches. The U^{235} fuel loading is 3.6 kilograms.

The reactor normally operates at a power level of 3000 kilowatts and has an average thermal flux in the core of 2.2×10^{13} neutrons$\cdot cm^{-2} \cdot sec^{-1}$.

The LITR is cooled by a closed cycle recirculating water system (Rupp and Cox, 1955) containing a total of approximately 10,000 gallons. The water enters the reactor tank at the top and is removed at the bottom, as shown in Figure 3. At the normal pumping rate of 1,200 gpm, the water is retained within the reactor core approximately 0.75 second.

A schematic flowsheet of the LITR cooling water system is shown in Figure 4. Water leaving the reactor is carried through an

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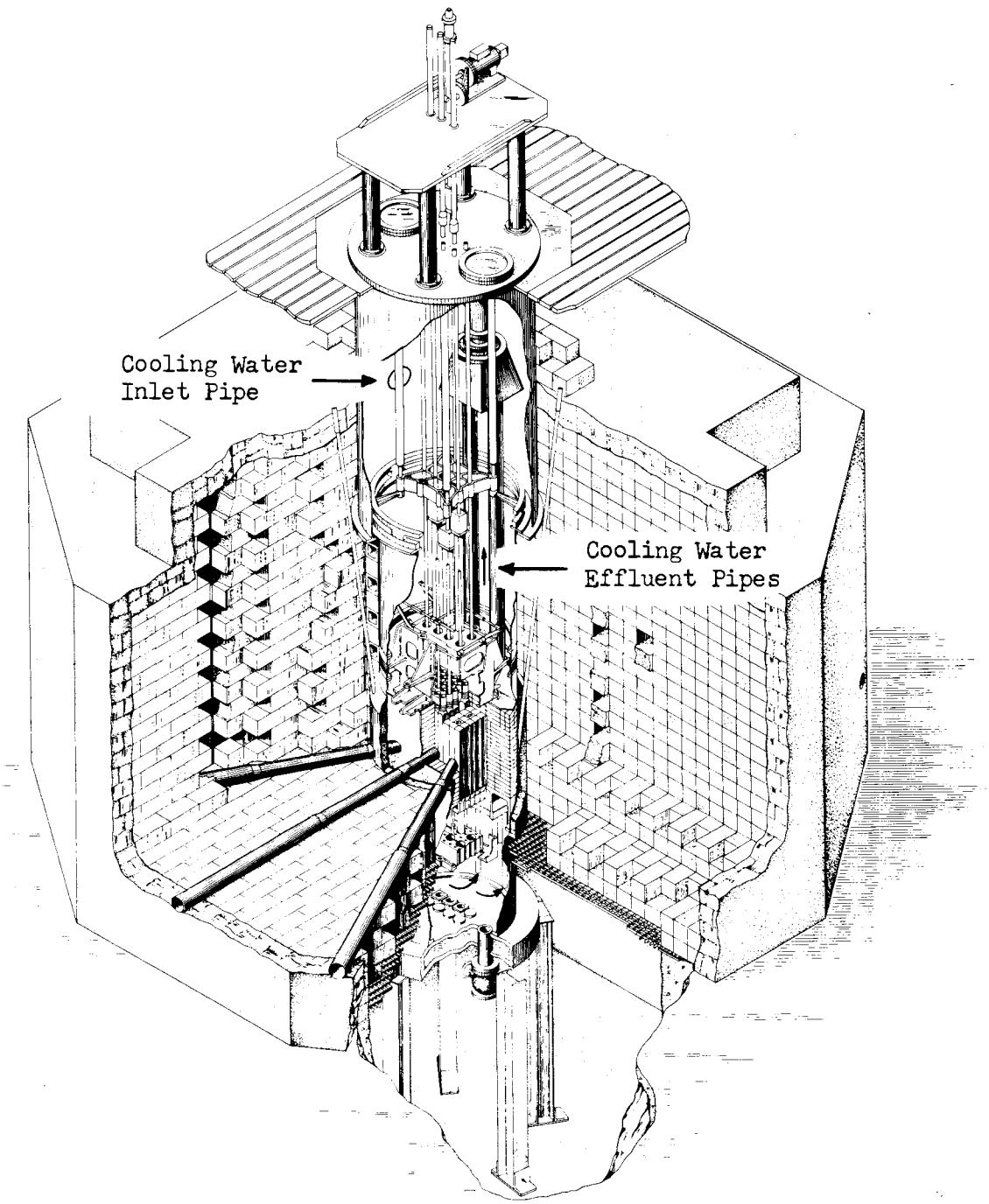

Fig. 3. Low Intensity Test Reactor: Core Vessel.

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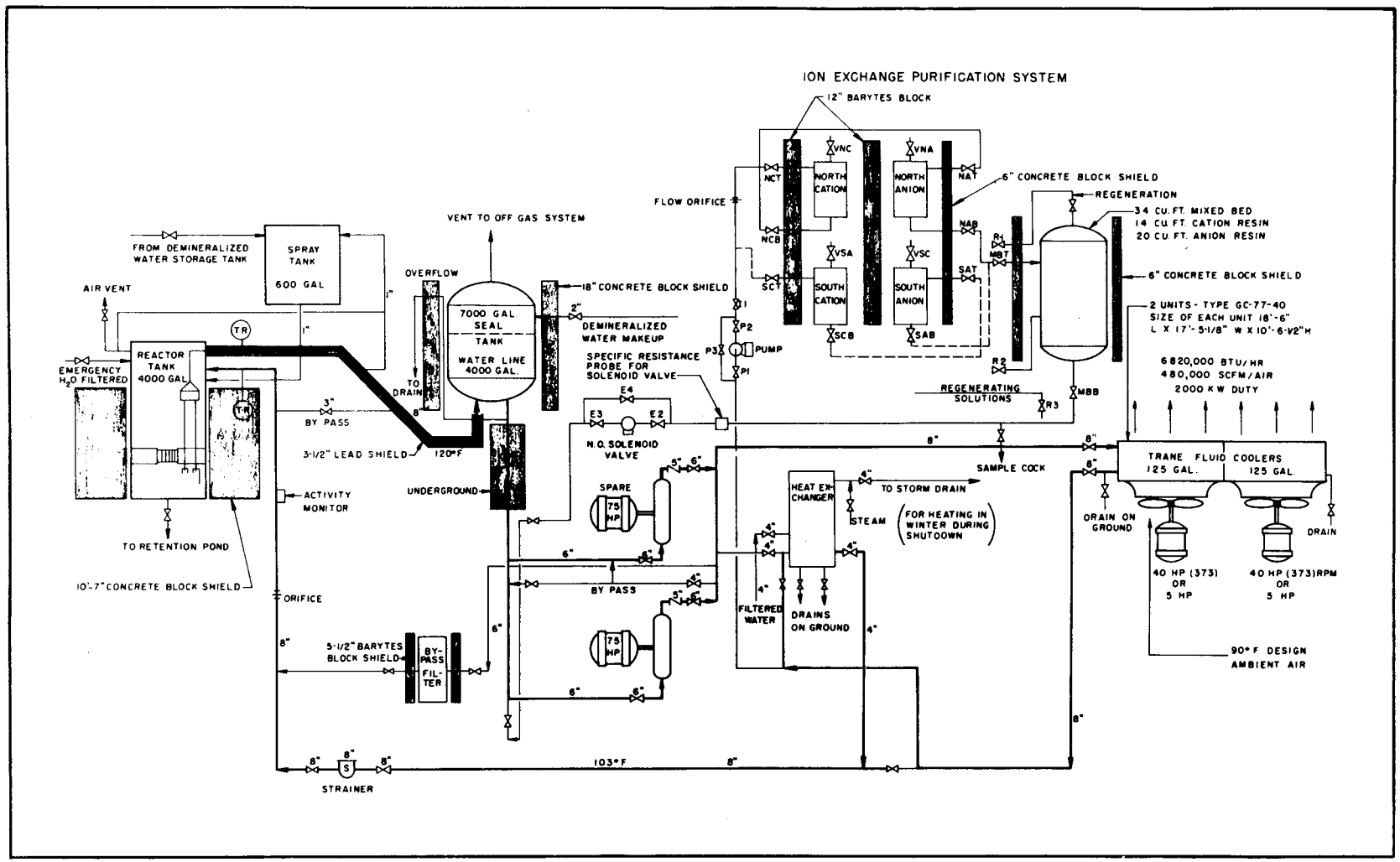

Fig. 4. Schematic Flowsheet: LITR Cooling Water System.

8 inch lead-shielded pipe to a 7000 gallon capacity concrete shielded seal tank. The water level in this tank is normally held at 4000 gallons. The primary purpose of this tank is to allow decay of the very short-lived radioactive nuclides, principally 7.4 s N^{16}, and to provide facilities for partial removal of dissolved gases in the water. From the seal tank the water is pumped to the air coolers and thereafter is returned to the reactor vessel for re-use.

Auxiliary facilities in the cooling system include a 600 gallon spray tank, a heat exchanger and an ion exchange purification system. The spray tank serves as an emergency supply of cooling water to prevent melting of the core in case of accidental loss of the water in the primary cooling system. The heat exchanger is used to prevent freezing of the water in the cooling system during periods of shut-down in winter months. It is also used as an auxiliary cooling unit during summer months. The ion exchange purification system, to be discussed in detail, maintains the quality of water in the over-all system.

Approximately 3% (≈40 gpm) of the cooling water flow is bypassed through the ion exchange purification system at all times. The system consists of three parts: a cation exchange bed, an anion exchange bed and a mixed resin ion exchange bed. The normal flow of water through the bypass system is as follows: the inlet water is passed through a cloth type clarifying filter (not shown), the cation column, the anion column, the mixed-bed column, a second clarifying filter (not shown) and then returned to the primary cooling system. There are two cation and two anion columns, one of each

type being in use at all times. There is only one mixed-bed column.

Some form of ion exchange purification system has been used to maintain the quality of the LITR cooling water since shortly after the reactor was built. However, the system was extensively modified during the summer of 1956 and the ion exchange units in the present system were first put into service on November 7, 1956. Specifically, they are as follows:

(a) two columns, 24 inches in diameter by 46 inches high, each containing 8 cubic feet of IR-120 cation resin,

(b) one column, 24 inches in diameter by 37 inches high, containing 6 cubic feet of IRA-410 anion resin and one column, 18 inches in diameter by 39 inches high, containing 3.5 cubic feet of IRA-410 anion resin,

(c) one mixed-bed column, 36 inches in diameter by 96 inches high, containing 20 cubic feet of IRA-410 anion resin and 14 cubic feet of IR-120 cation resin.

The cation units are shielded by a rectangular wall of 12 inch barytes block. The anion units are shielded by a 6 inch concrete block wall on three sides and share one wall of 12 inch barytes block with the cation units on the fourth side. The shielding serves also as weather protection. The units are covered by a conventional metal roof. A photograph of the cation and anion units, prior to construction of the shielding walls, is shown in Figure 5. One of the capped ports through which resin samples were obtained can be seen at the top of the nearest anion unit. The mixed-bed unit and its 6 inch concrete block shielding enclosure may be seen at the extreme left of the picture.

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Fig. 5. LITR Cooling Water System: Ion Exchange Units.

The purpose of the small cation and anion units is to protect the resin in the mixed-bed unit from radiation damage. Such damage apparently results in loss of resin efficiency and capacity. Of the two types of resin, the anion is apparently more susceptible to radiation damage (Rupp and Cox, 1955). Hence the cation bed precedes the anion in order to avoid as much exposure as possible to the latter.

Prior to re-entry into the primary cooling system, water leaving the ion exchange purification system passes through a solenoid valve. The valve is set to close automatically if the specific resistance of the water falls below 200,000 ohm-centimeters. In this way the valve serves to guard against inadvertent contamination of the primary cooling system with regenerating solutions and also serves to shut the bypass purification system off in the event of complete resin degeneration.

The frequency of regenerations of the cation and the anion exchangers is determined by their decontamination factor. Samples of their effluent are routinely counted for gross gamma activity. With freshly regenerated resin, the radioactivity in water passing through the columns is reduced by a factor of approximately 5 for the cation resin and by a factor of approximately 15 by the anion resin. Normally, the resin beds are removed from service when their effectiveness decreases approximately 50%. Figure 6 presents data on the range of typical removals of gross gamma activity from the cooling water by the ion exchangers during the period covered by this study.

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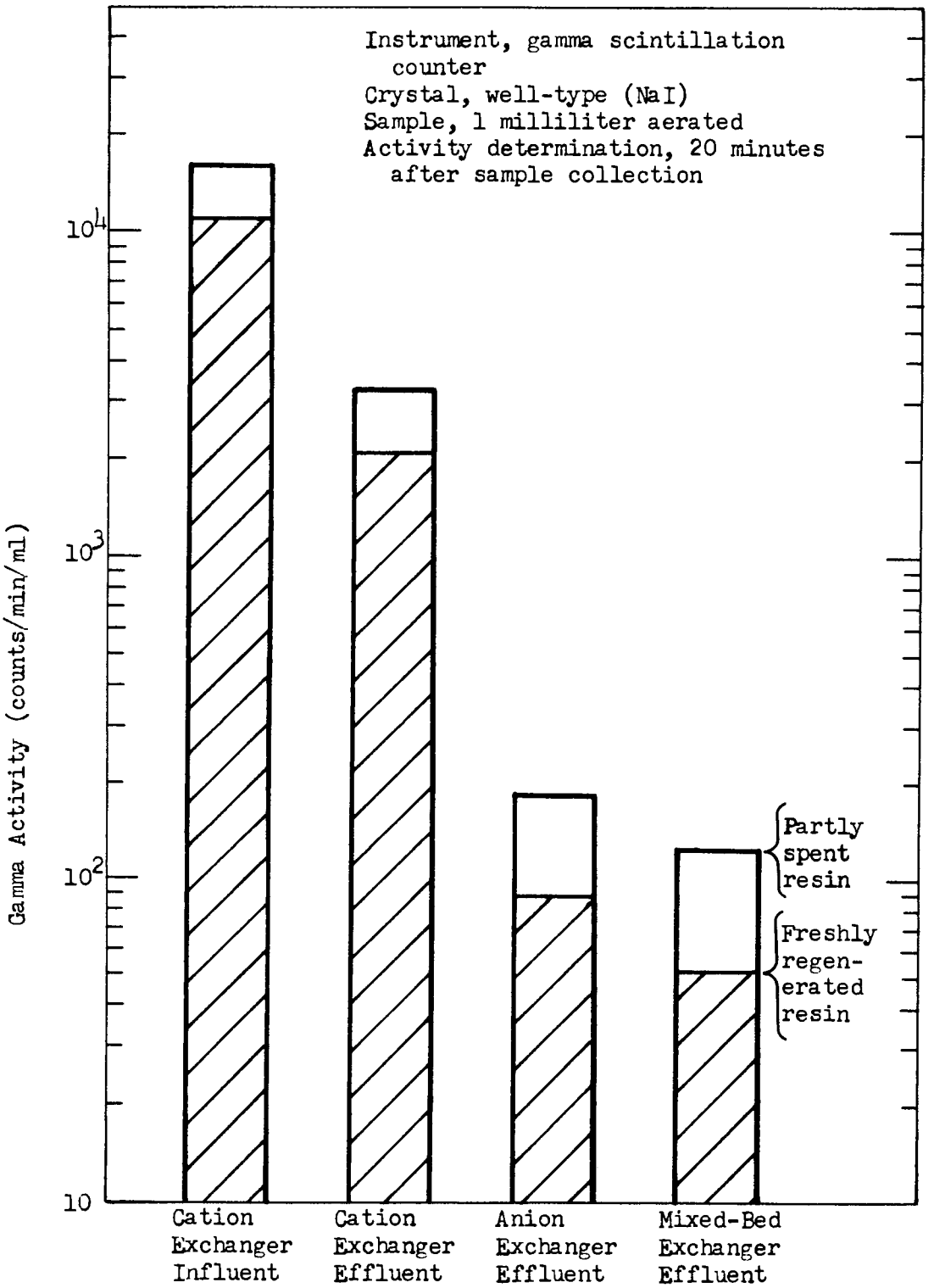

Fig. 6. Range of Gross Gamma Activity of LITR Cooling Water at Various Stages in Ion Exchanger Purification System.

When the specific resistance of the effluent from the mixed-bed unit drops to or near the resistance of the reactor cooling water, the unit is considered depleted. It is then isolated from the system and regenerated. Sulfuric acid and sodium hydroxide are used as regenerants. Radioactive regenerating solutions are stored to prevent release of radioactive material to the environment. The average specific resistance of the cooling water during the period of this investigation was 810,000 ohm-centimeters. The average life cycle of the mixed-bed unit was approximately four weeks, and the average number of gallons passing through the unit per cycle was approximately 1,500,000.

The mixed-bed exchanger was regenerated several times during the period covered by the experiments reported herein. The cation and anion exchangers were not regenerated during this period. However, as noted later in this report, both of the anion resin beds were used.

A somewhat similar demineralization system is used for the purification of the make-up water added to the LITR cooling system from time to time. The system consists of a cation exchanger followed by an anion exchanger and provides high purity water for other uses at the Laboratory as well as for LITR cooling system make-up.

CHAPTER V

ANALYTICAL TECHNIQUES

As previously stated, one of the principal objectives of this study was the identification of the radioactive nuclides in the cooling water at various stages in the system. The techniques employed are described below.

A. Radiochemical Assay

Gamma spectral and decay studies of samples as collected were inadequate for identification of the radionuclides present. Therefore, the water samples were chemically processed to concentrate and separate the radionuclides into groups whose identity could be established. The chemical procedure ultimately adopted represented a modification of that suggested by Lundell and Hoffman (1938).

Table I is a flow sheet of the chemical separation procedure. This procedure was found satisfactory for analysis of neutron activated water samples as well as most of those collected from the cooling water system. All samples to be processed were first aerated in a hood to remove dissolved radioactive gases. This procedure prevented unnecessary inhalation of radioactive gases by personnel processing the samples and also prevented colloidal carry down of radioactive gases during subsequent radiochemical procedures. After aeration, each sample was acidified with nitric acid and approximately three milligrams each of stable chlorine, bromine, iodine, copper,

TABLE I

Chemical Separation Scheme

1) Add an aliquot of the radioactive sample to a 50 milliliter centrifuge tube. Aerate to remove radioactive gases. Acidify with HNO_3 and add Cl, Br, I, Cu, As, Fe, Y, Co, Mn, Zn, Ba, Sr, Ca, Na, and K carriers. Precipitate Cl, Br, and I with slight excess of Ag. Heat to coagulate the precipitate. Centrifuge.

2) Precipitate: AgCl, AgBr, AgI. Wash with water, filter, mount and count. Designate as the SILVER GROUP.

3) Supernate: Adjust acidity to 0.5 N HCl. Saturate with H_2S. Centrifuge.

4) Precipitate: $CuS + As_2S_3$. Wash with water, filter, mount and count. Designate as the ACID SULFIDE GROUP.

5) Supernate: Transfer to beaker. Add 1 ml. conc. HCl. Boil until solution is clear. Oxidize Fe^{++} to Fe^{+++} by boiling with HNO_3. Cool solution and make 1:1 with conc. HCl. Neutralize with conc. NH_4OH to precipitate hydroxide. Centrifuge.

6) Precipitate: Fe + Y hydroxides. Wash with water, filter, mount and count. Designate as the HYDROXIDE GROUP.

7) Supernate: Saturate with H_2S. Centrifuge.

8) Precipitate: CoS, MnS, ZnS. Wash with water, filter, mount and count. Designate as the BASIC SULFIDE GROUP.

9) Supernate: Acidify solution with HCl. Boil to remove H_2S. Add $(NH_4)_2H(PO_4)$ and an excess of NH_4OH. Digest. Centrifuge.

10) Precipitate: Ca, Ba, and Sr Phosphates. Wash with water containing a few drops of NH_4OH. Filter, mount and count. Designate as the ALKALINE EARTH GROUP.

11) Supernate: Na + soluble ions. Transfer aliquot to culture tube for counting. Designate as the SOLUBLE GROUP.

arsenic, iron, yttrium, cobalt, manganese, zinc, barium, strontium, calcium, sodium and potassium were added to serve as isotopic or non-isotopic carriers for the radioactive nuclides present. The resulting mixture was then processed to yield precipitates of silver chloride, bromide and iodide (Silver Group), copper and arsenic sulfide (Acid Sulfide Group), iron and yttrium hydroxide (Hydroxide Group), cobalt, manganese and zinc sulfide (Basic Sulfide Group), calcium, barium and strontium phosphate (Alkaline Earth Group) and a soluble fraction (Soluble Group) containing those radionuclides not carried by the stable carrier elements used.

Since bromine and iodine proved to be important radioactive nuclides, silver was added in excess to insure their complete precipitation in the Silver Group. The excess silver was subsequently precipitated as the chloride upon acidification of the sample for the acid sulfide step. However, the radioactive material contained in this additional silver precipitate was found to be negligible.

Complete separation of the various radionuclides in a given sample is not always readily possible. The use of carriers constitutes the main control. However, in certain instances, specific steps must be employed to control observed contamination. For example, the precipitation of the Hydroxide Group in the presence of a high concentration of ammonium chloride was found necessary to prevent manganese contamination of this same group.

It is possible, of course, to further separate each of the given chemical groups where necessary for positive identification

in questionable cases. In several of the samples discussed later in this report such techniques were employed.

Because of the time required for chemical processing of samples, it would be anticipated that the very short-lived radionuclides commonly reported in cooling water would not be detected in these studies.

B. Radioactivity Assay

After each chemical group separation the sample was assayed by gross beta or gamma decay studies, beta absorption determinations and/or gamma spectral analyses. An interpretation of these data, together with knowledge of the chemistry of the particular group, was used to identify the radionuclides in the sample (Reynolds, 1955; Kahn and Lyon, 1953).

Gross gamma decay studies were made utilizing a well-type scintillation detector coupled to a Model No. 101B research scaler manufactured by the Atomic Instrument Company of Cambridge, Massachusetts. Possible choices of scaling factors ranged from 16 to 1024. Coincidence loss was one percent per 10^5 counts per minute. A linear amplifier was furnished as an integral part of the scaler. High voltage was obtained from a power supply also manufactured by the Atomic Instrument Company, Model No. 312. The detector was a NaI(Tl) crystal two inches in length and two inches in diameter with a central well 1.5 inches deep by 0.86 inch in diameter. The well contained an aluminum liner which acted as an absorber for

the beta radiation emitted by the sample being analyzed. A three inch lead shield surrounded the crystal detector and sample being analyzed (Brooksbank, 1956).

Gross beta decay studies were made using a thin mica-window Geiger-Mueller detector, type TGC-2, manufactured by Tracerlab, Inc., of Boston, Massachusetts. The detector was coupled to a scaler, with scale of 64, manufactured by the Nuclear Instrument and Chemical Corporation of Chicago, Illinois. The high-voltage supply was contained in the same chassis. Coincidence loss was approximately one half percent per 10^3 counts per minute. A two inch cylindrical lead shield surrounded the detector and sample being analyzed. Beta absorption studies were made utilizing this same instrumentation and calibrated aluminum absorbers made locally.

Gamma spectral data were obtained using both a single channel scintillation scanning spectrometer and a multichannel spectrometer. In the single channel unit only a narrow band of pulses in the gamma spectrum was viewed at one time through an electronic slit. The slit was variable and could be moved to cover the entire spectrum. Automatic scanning of the spectrum was accomplished by means of a potentiometer driven by the chart drive of a Brown recorder. The input signal to the recorder was from a logarithmic count-rate meter fed by a discriminator circuit (Brooksbank, 1956). The scintillation detector was of the well-type similar to that previously described as being used in the gross gamma decay studies.

For samples containing a gross gamma activity of approximately 10,000 counts per minute or more, the single channel spectrometer was operated so that the complete energy range was scanned in fifteen minutes. For samples containing approximately 5,000 counts per minute or less the scanning time was one hour. A coarse gain control permitted selection of the energy range to be scanned. The two settings most commonly used in these studies were 32 and 8, which corresponded to energy ranges of 0 to 0.5 Mev and 0 to 2 Mev, respectively.

The multichannel gamma scintillation spectrometer was of the 20 channel type. The instrument, Model 520, Twenty-Channel Pulse Height Analyzer, was manufactured by the Atomic Instrument Company. The detector was of the well-type previously described. The pulses were differentiated and fed into twenty separate scalers, each having a scale factor of 16. Controls were available for adjusting the channel widths and selecting the portion of the total spectrum to be recorded. Suitable adjustment of the controls permitted recording the total count for a given time in each of 20 adjacent channels. Five settings of 20 channels each were necessary to cover the energy range 0.08 to 2.0 Mev. This range was most commonly used in the determinations reported herein.

Three radioactive nuclides of known gamma energies were used for daily calibration of both spectrometers. These nuclides and their characteristic gamma energies were: (a) 48 d Hg^{203} (0.279 Mev); (b) 30 y Cs^{137} + Ba^{137} (0.662 Mev); (c) the positron emitter 2.6 y Na^{22} (1.28 Mev).

In all counting determinations, numerical corrections were made to account for background radiation and coincidence losses.

Samples for beta studies were placed on cellophane-covered watch glasses fastened to cardboard mounts. Samples for gamma studies were mounted in 10 x 75 millimeter Pyrex culture tubes. These culture tubes fit down into the aluminum liner of the well-type scintillation crystal, permitting each sample to be essentially surrounded by the detector crystal.

CHAPTER VI

RADIOACTIVE NUCLIDES PRESENT IN LITR COOLING WATER

In order to determine the radioactive nuclides in the cooling water, to evaluate the performance of the bypass ion exchange purification system and to gain knowledge as to the source of the nuclides in the water, it was necessary to sample the cooling system at several points. In the discussion to follow each such sample will be considered separately.

During routine operation, the LITR is shut down one day each week for fuel loading, changes in experimental setups and other necessary alterations. To avoid non-equilibrium conditions all samples were collected several days after the reactor had been brought back to full power. Many samples were collected. For purposes of this report, data for only a single sample, typical of those collected at each point, are presented.

A. Make-Up Water (Analysis for Stable Elements)

One of the possible sources of radioactive nuclides in cooling water is the activation of dissolved solids present in the original water making up the system and in the water added to the system from time to time to replace losses. Hence, as an initial step in the study of the radioactive nuclides present in the cooling water of the LITR, it was deemed important to determine the purity of the demineralized make-up water. The technique employed for this purpose

was neutron activation analysis (Leddicotte and Reynolds, 1951; Smales, 1953). This choice was made for two reasons: (a) those elements which would lead to induced activities were of primary importance; (b) the necessity for an extremely sensitive technique. Spectrographic analysis had revealed, for example, only that the water contained 0.02 ppm aluminum and a faint trace of magnesium.

The sample of make-up water collected for these studies had a specific resistance of 1,800,000 ohm-centimeters and a pH of 6.2. A 40 milliliter aliquot was placed in a polyethylene bottle and irradiated in Hole 10 of the ORNL Graphite Reactor. Hole 10 is a water cooled irradiation facility having a thermal flux of approximately 6.5×10^{11} neutrons$\cdot$cm$^{-2}\cdot$sec^{-1}. The time of irradiation was chosen as 16 hours since it was known from previous work that radioactive nuclides such as Cu^{64} and Na^{24} could be expected in cooling water. Such an irradiation period would show the presence of these particular nuclides, should stable copper or sodium occur in the make-up water.

The activated sample was chemically separated by the method previously outlined. Gamma spectral studies of the Silver Group utilizing the single channel spectrometer were inconclusive. (The 20 channel gamma scintillation spectrometer was not available during study of this initial sample.) However, beta decay and absorption studies revealed the presence of nuclides with half lives of approximately 36 and 5 hours and possessing beta energies of approximately 0.5 and 2.0 Mev. (See Figures 7 and 8). These

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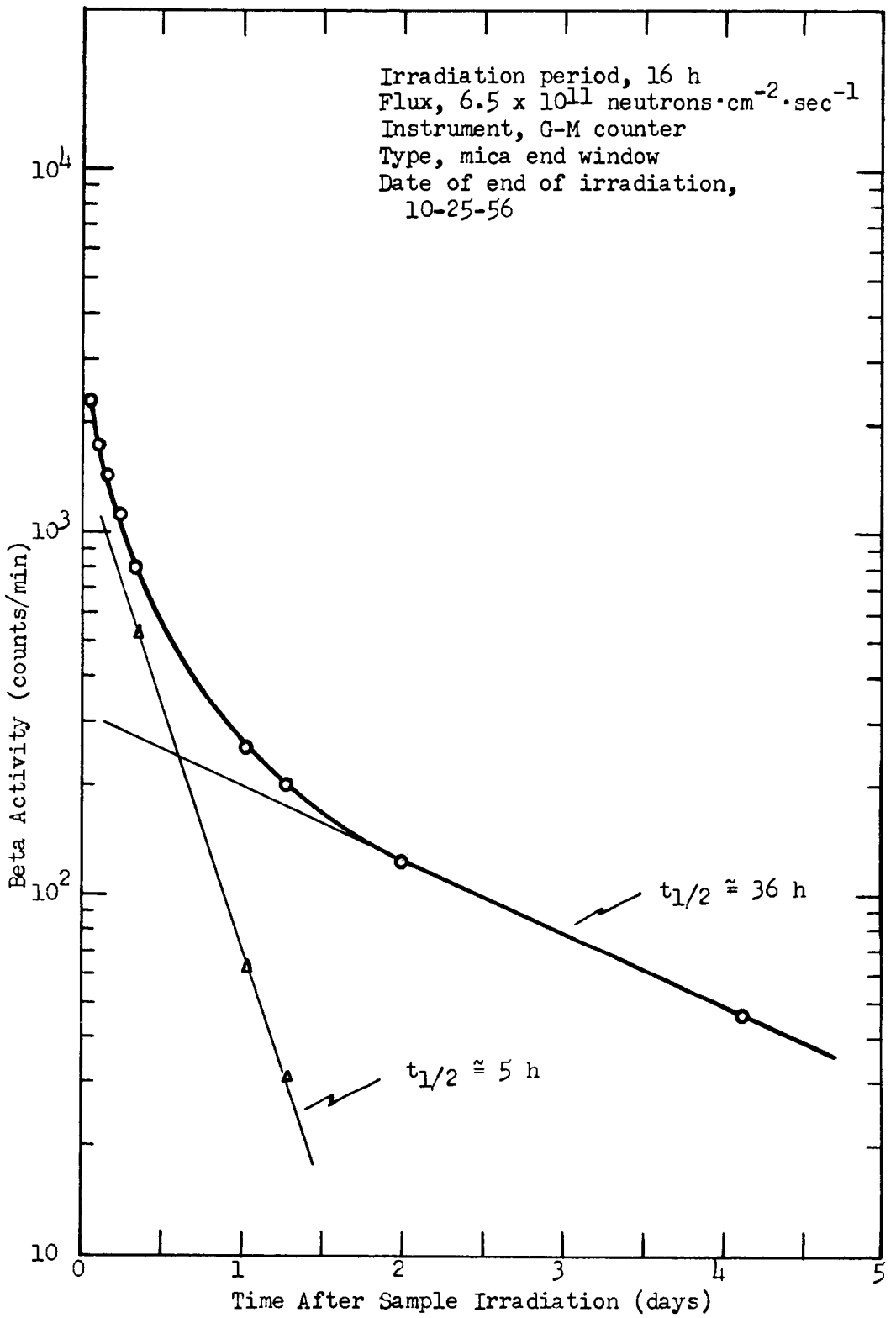

Fig. 7. Gross Beta Decay for Neutron Irradiated LITR Make-Up Water: Silver Group.

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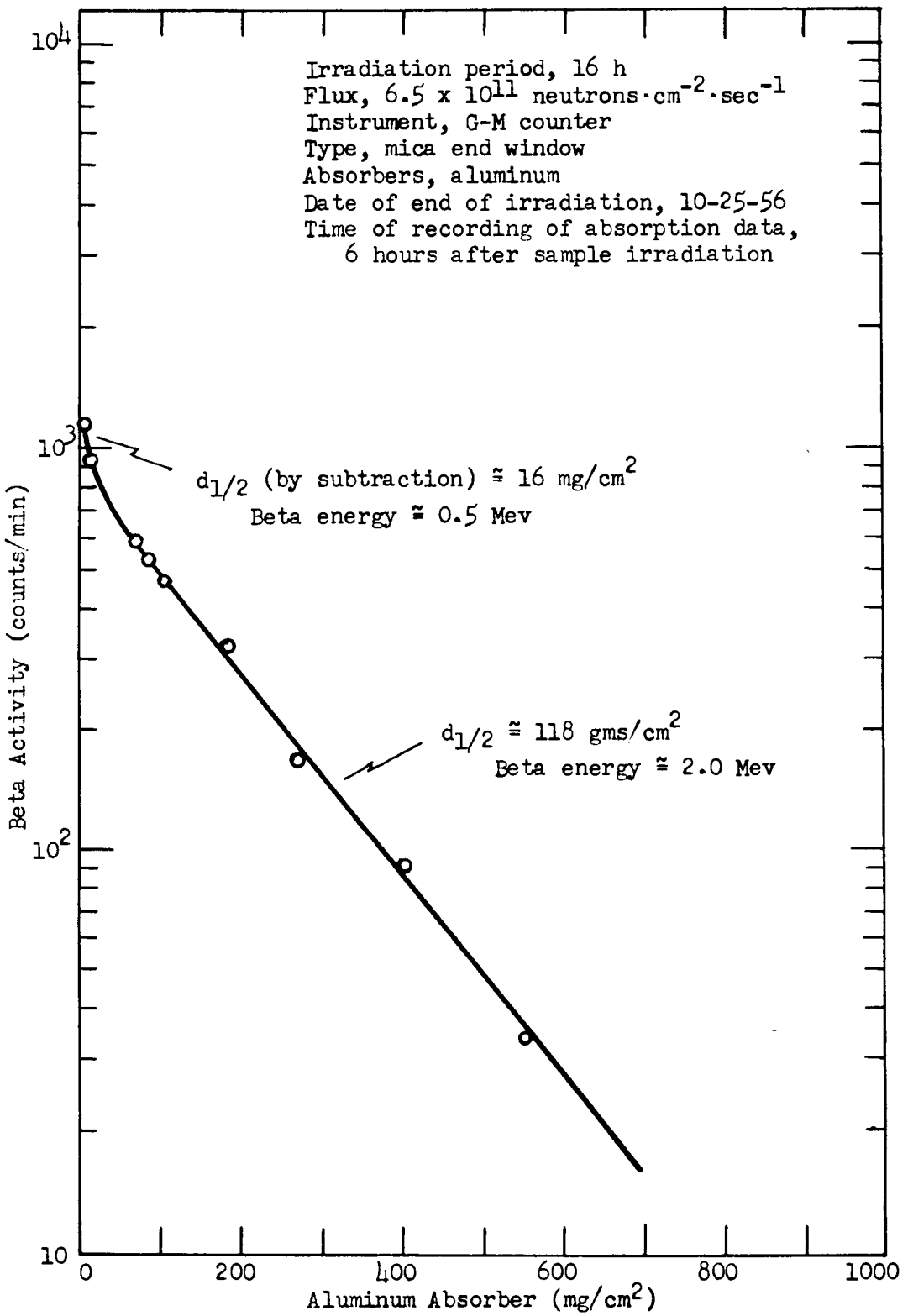

Fig. 8. Beta Absorption Data for Neutron Irradiated LITR Make-Up Water: Silver Group.

properties are characteristic of 35.9 h Br^{82} and 4.6 h Br^{80m} + Br^{80} A gross gamma decay study of the Silver Group from a similar water sample irradiated for 20 minutes confirmed the presence of 18 m Br^{80}. All three of these nuclides originate by the (n,γ) reaction on stable bromine.

Gamma decay and single channel spectral studies of the remainder of the chemical groups obtained from this sample proved adequate for the determination of the more prominent gamma emitting radionuclides present. Figure 9, the spectral data for the Acid Sulfide Group, shows the characteristic 0.51 Mev annihilation gamma peak for the positron emitter Cu^{64}. Gamma energies on the spectral curves are listed as experimentally determined. Due to instrument variations and calibration errors, energies for a given nuclide may vary from sample to sample. The 1.05 Mev peak is a result of "pile up". Peaks due to "pile up" are quite common in gamma spectral studies utilizing the well-type crystal detector. They may be attributed to coincidence summing. A "pile up" peak occurs when two photons appear in the crystal detector simultaneously, resulting in a photopeak representing their combined energies rather than the energy of either single photon. The particular peak here is due to the summing of two annihilation photons (Siegbahn, 1955).

The gamma spectral data combined with the half life data, Figure 10, showing the presence of an approximately 13 h component, confirm the identification of Cu^{64}. The 2.7 d half life component is probably indicative of the radionuclide Sb^{122}.

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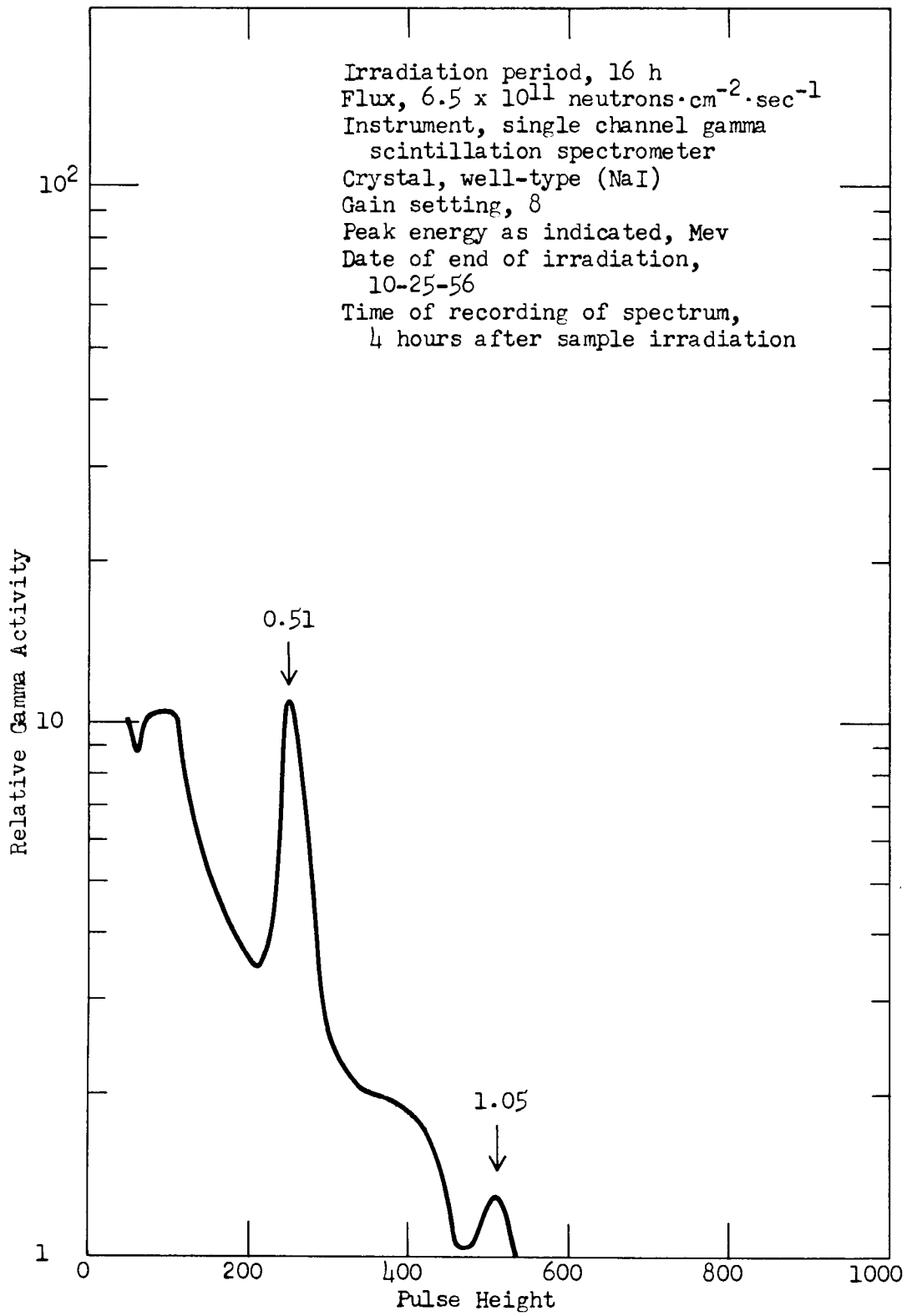

Fig. 9. Gamma Spectral Data for Neutron Irradiated LITR Make-Up Water: Acid Sulfide Group.

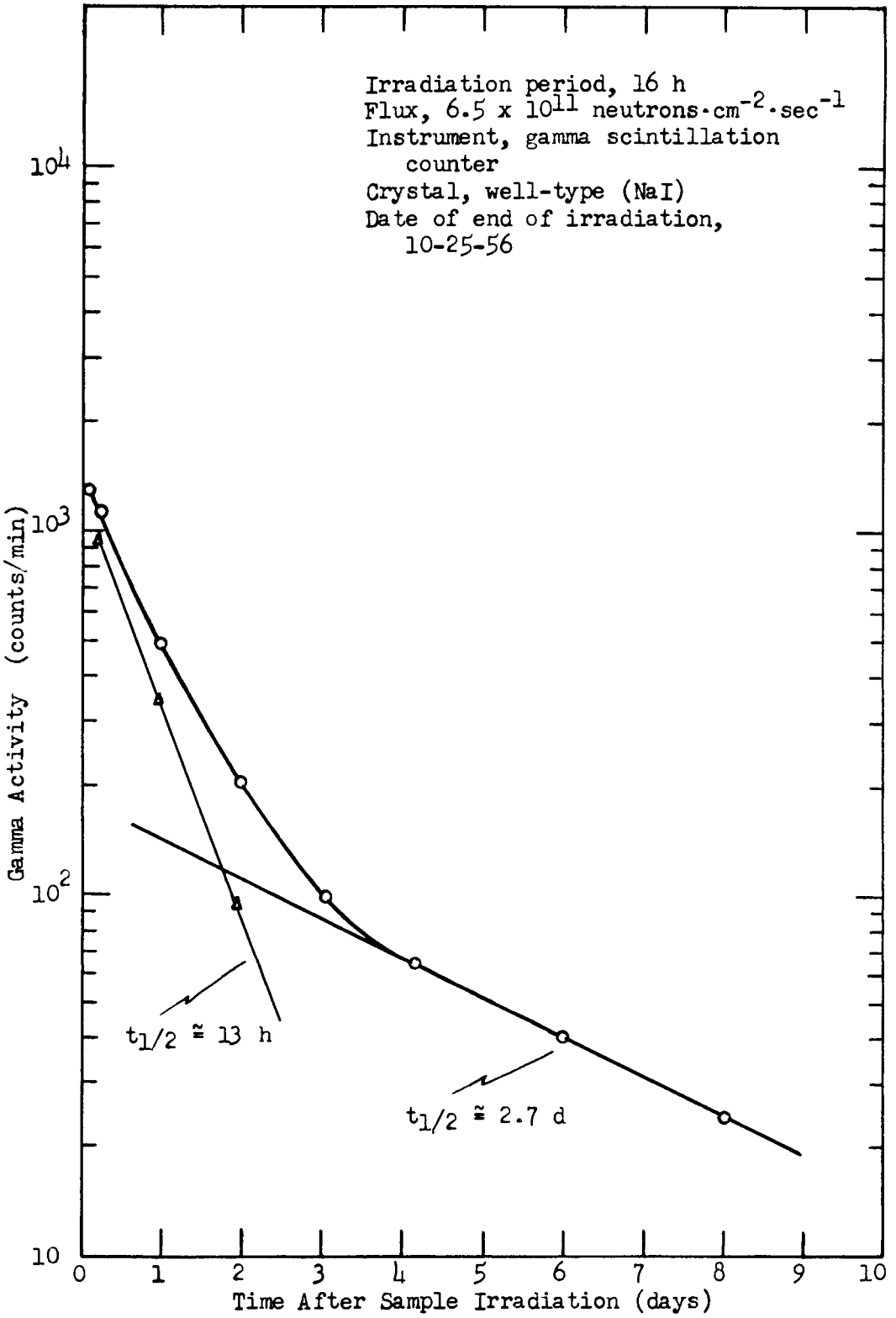

Fig. 10. Gross Gamma Decay for Neutron Irradiated LITR Make-Up Water: Acid Sulfide Group.

Gamma spectral and decay studies of the Hydroxide and Alkaline Earth Groups revealed the possible presence of 1.87 h F^{18} in the water. To confirm these findings, a second sample of make-up water was irradiated and passed through a small laboratory cation exchanger. Stable fluoride carrier was then added to the exchanger effluent and precipitated as lanthanum fluoride. Gamma spectral and decay data for the precipitate are shown in Figures 11 and 12. These data are indicative of F^{18}.

Inasmuch as F^{18} would not be produced in neutron irradiated water by the common (n,γ) reaction, some time was devoted to determining its source. The addition of stable fluoride in the form of hydrofluoric acid to similar water samples prior to irradiation showed the source of the F^{18} was not stable fluorine, i.e. by the reaction $F^{19}(n,2n)F^{18}$. After additional study, it was finally concluded that the most likely source of the F^{18} was the (p,n) reaction on O^{18}, the source of the protons being "knock-on" protons produced from the water by fast fission neutrons. The cross section for the $O^{18}(p,n)F^{18}$ reaction for protons of 4 Mev energy has been reported to be 0.2 barns, the threshold for the reaction being 2.56 Mev with a resonance maximum at 3.55 Mev (DuBridge, et al., 1938). Heath (1956) had previously reported the finding of F^{18} in MTR cooling water and had reached the same conclusion as to its source.

Gamma spectral and decay studies of the Basic Sulfide Group are shown in Figures 13 and 14. The 0.82 Mev gamma peak is characteristic of 2.58 h Mn^{56}. The 0.51 Mev gamma peak again suggests the presence of 1.87 h F^{18}.

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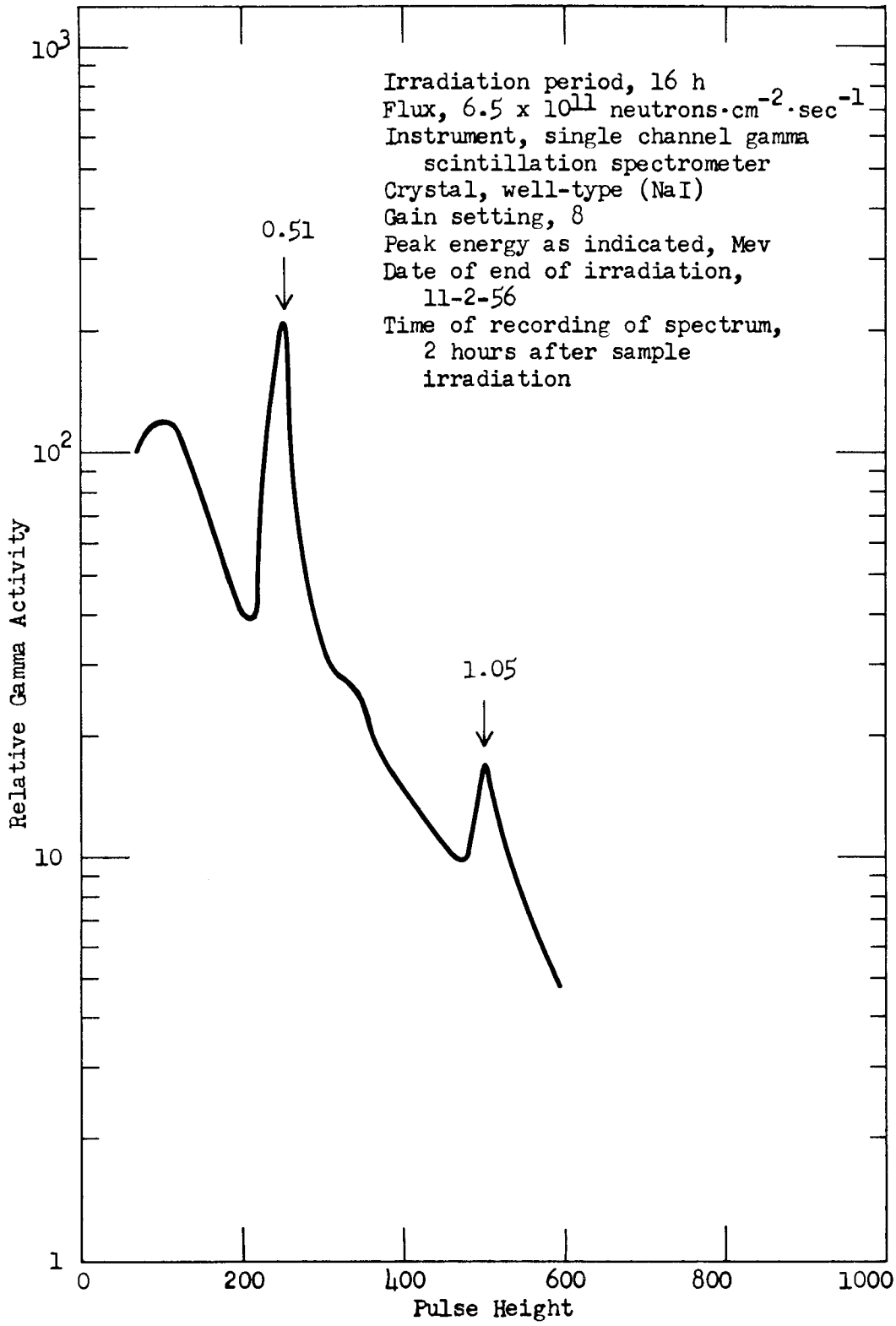

Fig. 11. Gamma Spectral Data for Neutron Irradiated LITR Make-Up Water: LaF_3 Precipitate.

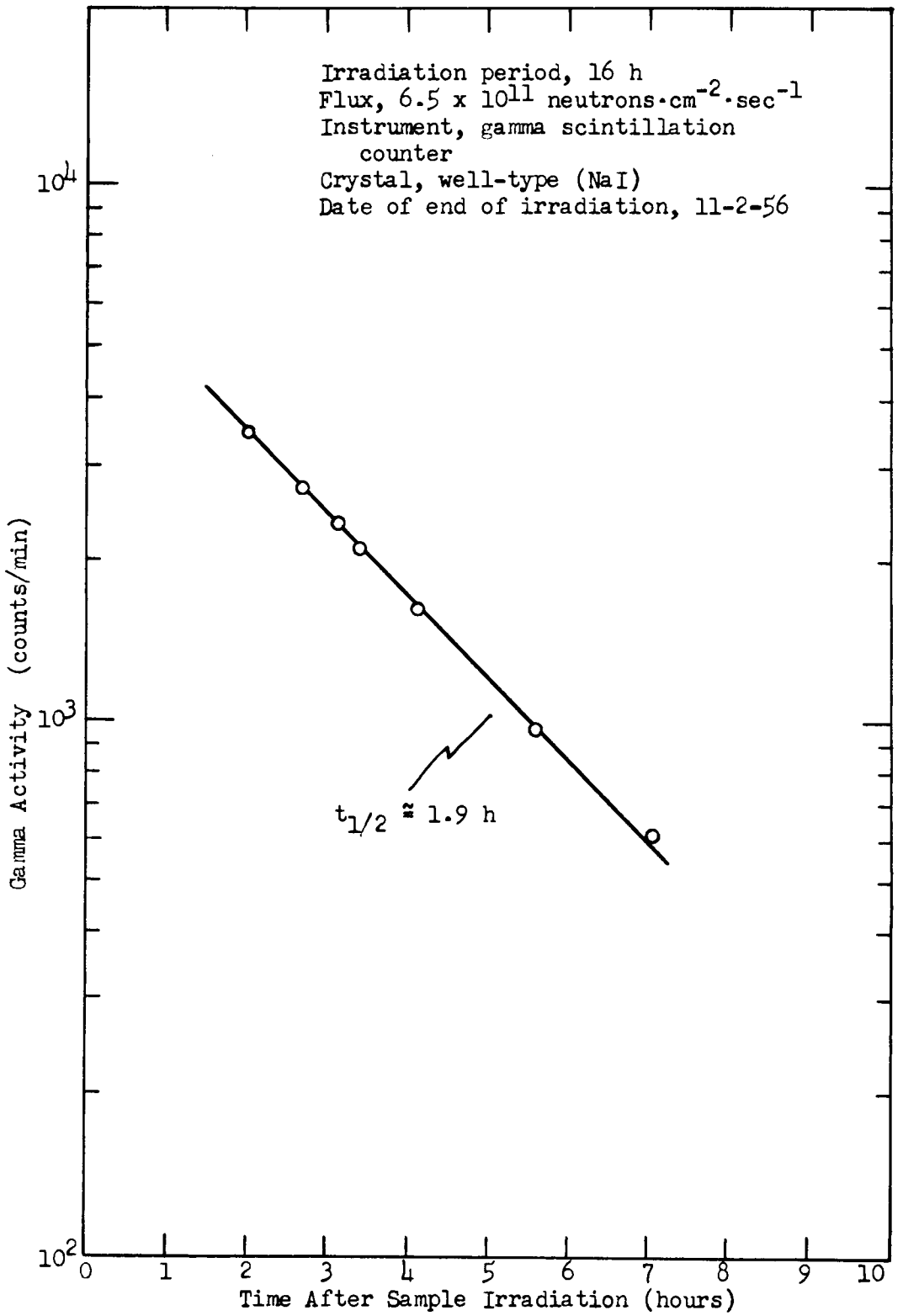

Fig. 12. Gross Gamma Decay for Neutron Irradiated LITR Make-Up Water: LaF_3 Precipitate.

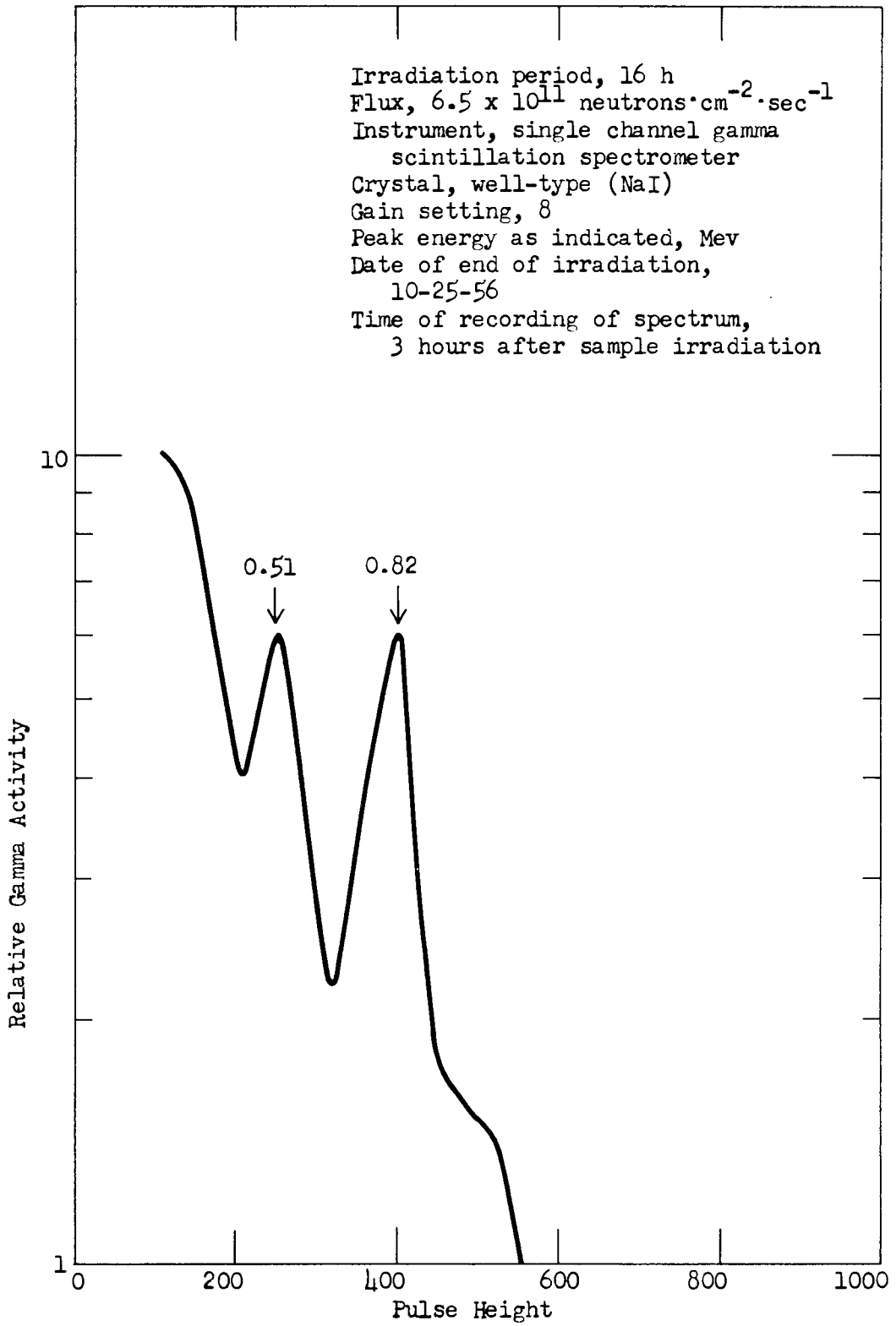

Fig. 13. Gamma Spectral Data for Neutron Irradiated LITR Make-Up Water: Basic Sulfide Group.

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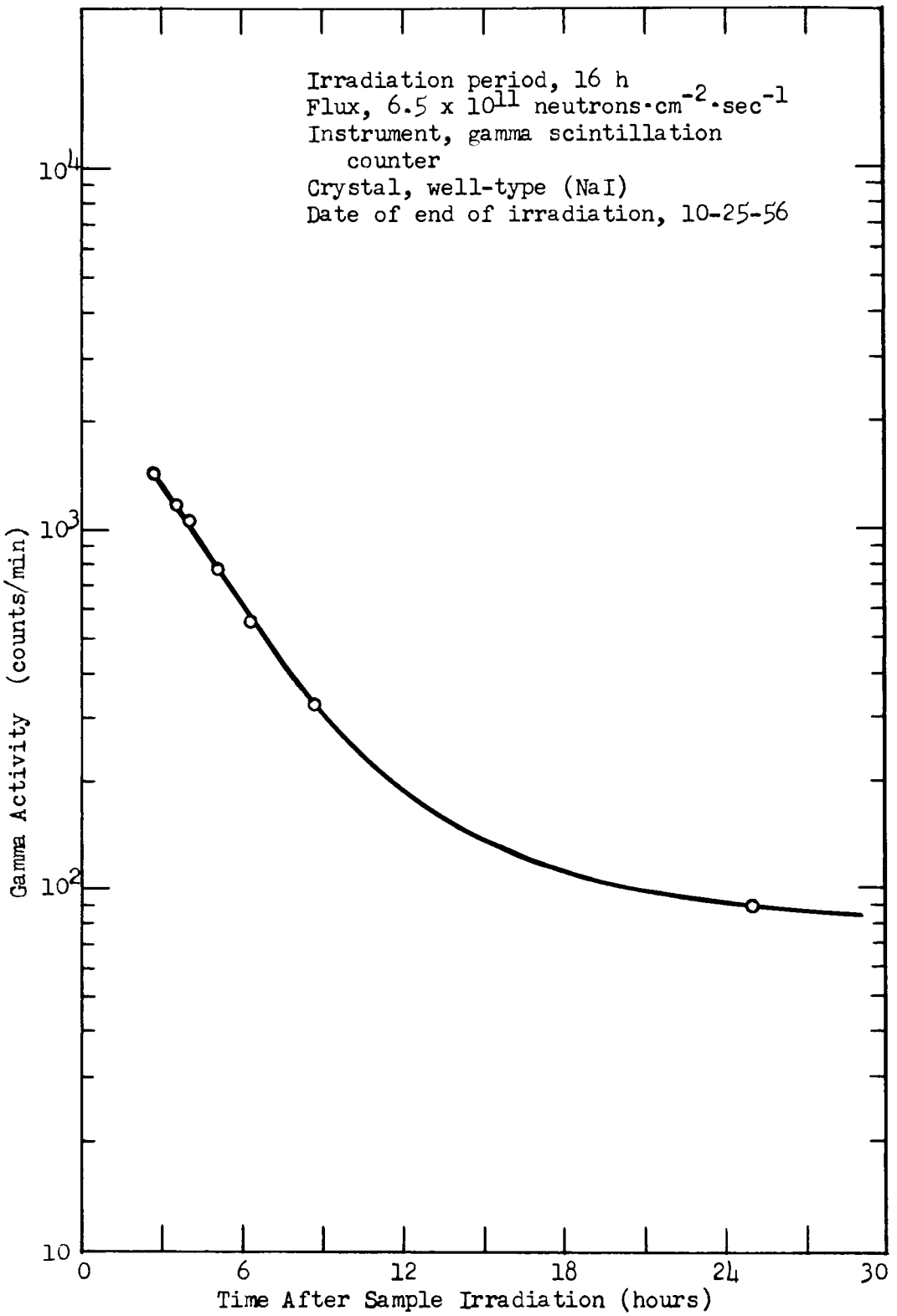

Fig. 14. Gross Gamma Decay for Neutron Irradiated LITR Make-Up Water: Basic Sulfide Group.

Gamma spectral and decay studies of the Soluble Group are shown in Figures 15 and 16. The 1.34 and 2.75 Mev gamma peaks are characteristic of 15.0 h Na^{24}.

A summary of the stable element concentration in the make-up water as determined by neutron activation analysis is given in Table II. From the known irradiation period and thermal neutron flux, and the half life, activation cross section and experimentally measured activity of each of the induced radionuclides in the irradiated sample, the concentration (ppm) of the stable, source element was calculated. An average chemical yield of 75%, and beta and gamma counting efficiencies of 10% and 33%, respectively, were assumed in making the calculations. The data presented in Table II therefore illustrate orders of magnitude only.

B. Cation Exchanger Influent

Inasmuch as the overall objective of this investigation was to determine the radioactive nuclides present in the cooling water and to evaluate the efficiency of the ion exchange purification system, the point chosen for initial sampling was the influent line to the cation exchanger. It was believed that such a sample would be representative of the cooling water in general as well as provide initial conditions for evaluation of the purification system. The time required for water leaving the reactor vessel to reach this initial sampling point was 5 to 10 minutes. As previously pointed out, additional delays during chemical processing of the samples

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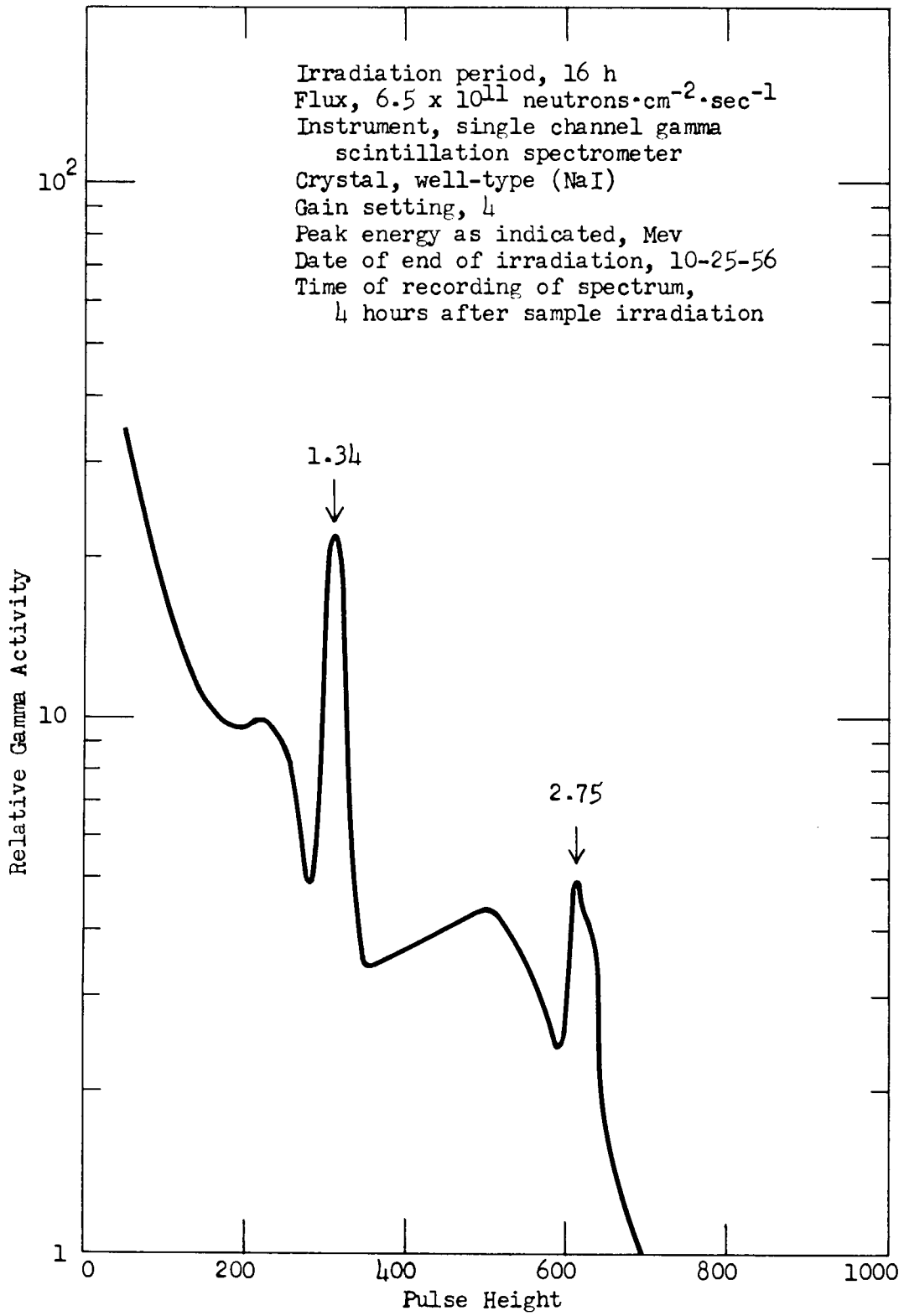

Fig. 15. Gamma Spectral Data for Neutron Irradiated LITR Make-Up Water: Soluble Group

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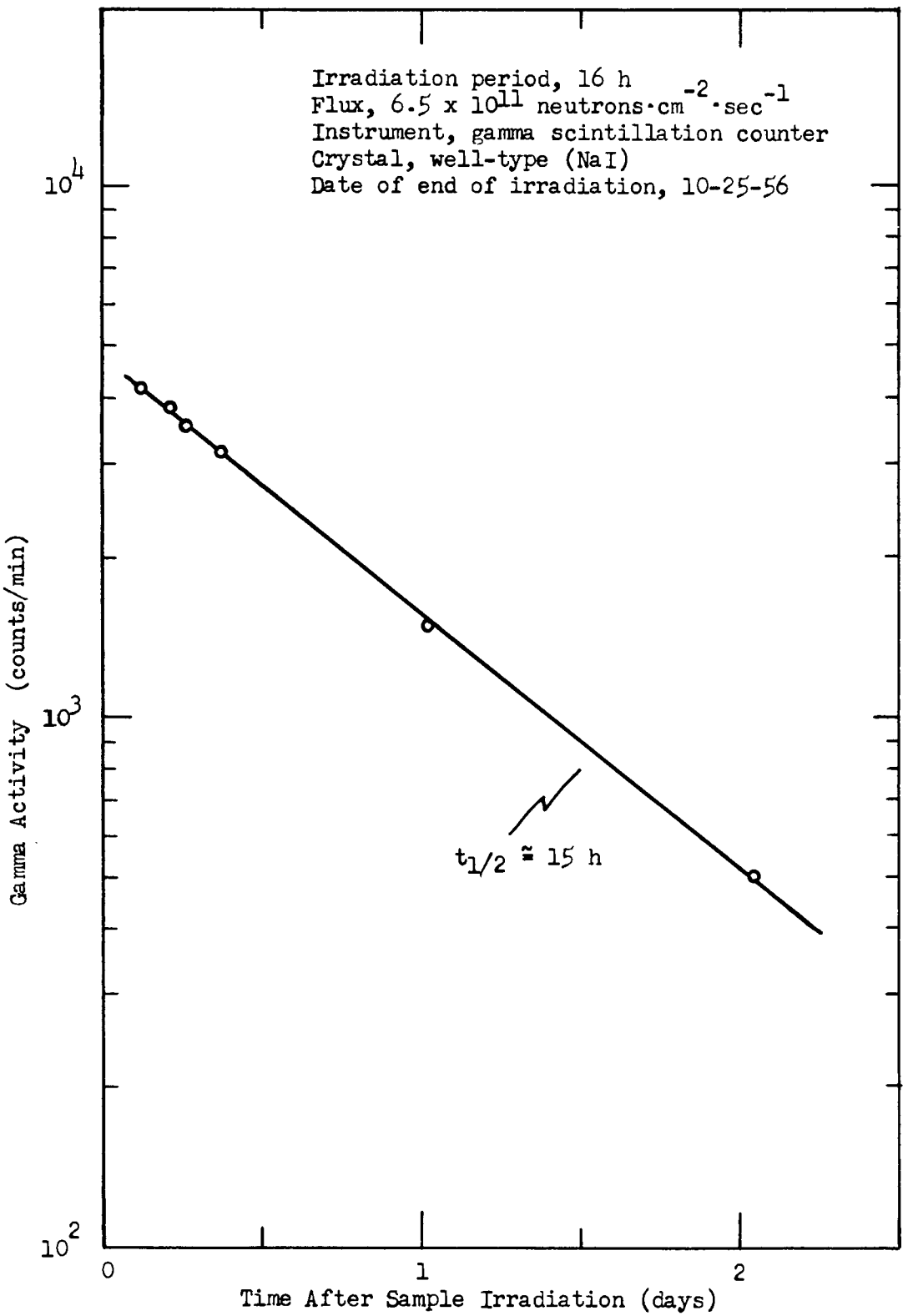

Fig. 16. Gross Gamma Decay for Neutron Irradiated LITR Make-Up Water: Soluble Group.

TABLE II

Concentrations of Stable Elements in LITR Make-Up Water as Determined by Neutron Activation Analysis

Irradiation period, 16 h
Flux, 6.5×10^{11} neutrons·cm^{-2}·sec^{-1}
Date of sample collection, 10-24-56
Date of end of irradiation, 10-25-56

Chemical Group	Induced Radio-nuclide	Half Life	Estimated Concentration (μc/ml)*	Stable Element Source	Estimated Stable Element Concentration (ppm)
Silver	Br^{80} Br^{80m} Br^{82}	18 m 4.6 h 35.9 h	5×10^{-5} } 8×10^{-4}	Bromine	2×10^{-3}
Acid Sulfide	Cu^{64}	12.8 h	4×10^{-5}	Copper	2×10^{-4}
Hydroxide and Alkaline Earth	F^{18}	1.87 h	3×10^{-4}	Oxygen	---
Basic Sulfide	Mn^{56} F^{18}	2.58 h 1.87 h	1×10^{-4}	Manganese	4×10^{-5}
Soluble	Na^{24}	15.0 h	5×10^{-3}	Sodium	4×10^{-2}

* At end of irradiation.

prevented identification of the very short-lived radionuclides which may have been present.

A sample volume of 140 milliliters was collected and chemically separated as previously outlined. For spectral studies, the primary instrument used was the 20 channel gamma scintillation spectrometer, the single channel unit serving only as a supplementary unit for "spot" checks.

Gamma spectral and decay data for the Silver Group are shown in Figures 17 and 18. The spectral data show that 21 h I^{133}, 8.05 d I^{131} and their daughter products constitute a major portion of this chemical group. The presence of I^{133} is characterized by the prominent 0.53 Mev gamma peak and the lesser 0.87 and 1.31 peaks as shown in Curve B of Figure 17. Iodine-131 is characterized by the prominent 0.37 and 0.64 Mev peaks in Curve C of the same figure, as well as the approximately 8 d half life shown in Figure 18. The 0.08 Mev peak is representative of 5.27 d Xe^{133} and its daughter Cs^{133m} as well as 4.8×10^{10} s Xe^{131m1}, daughter products of I^{133} and I^{131}, respectively.

The prominent 0.25 Mev peak in Curves A and B is characteristic of 9.1 h Xe^{135} + Cs^{135m}, which strongly suggests the presence of its parent 6.7 h I^{135}. Iodine-135 is confirmed by the presence in Curve B of peaks near its characteristic gamma-energies of 0.84, 1.13, 1.28, 1.46 and 1.72 Mev (Heath, 1956).

The rapid initial gross gamma decay of the sample as shown in Figure 18 strongly suggests also the presence of 52 m I^{134}. However, its presence would be difficult to confirm by the spectral data.

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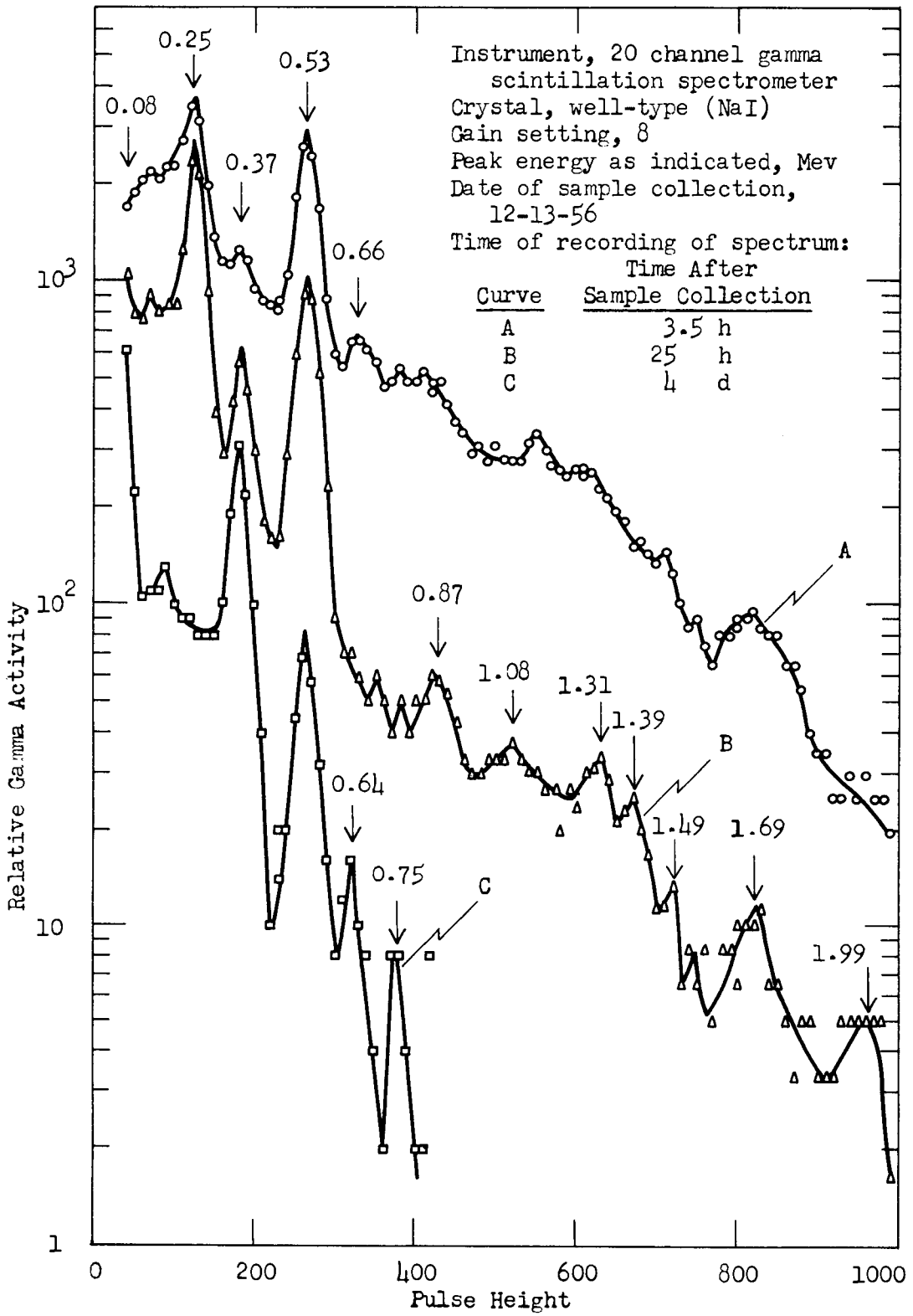

Fig. 17. Gamma Spectral Data for Influent to LITR Cation Exchanger: Silver Group.

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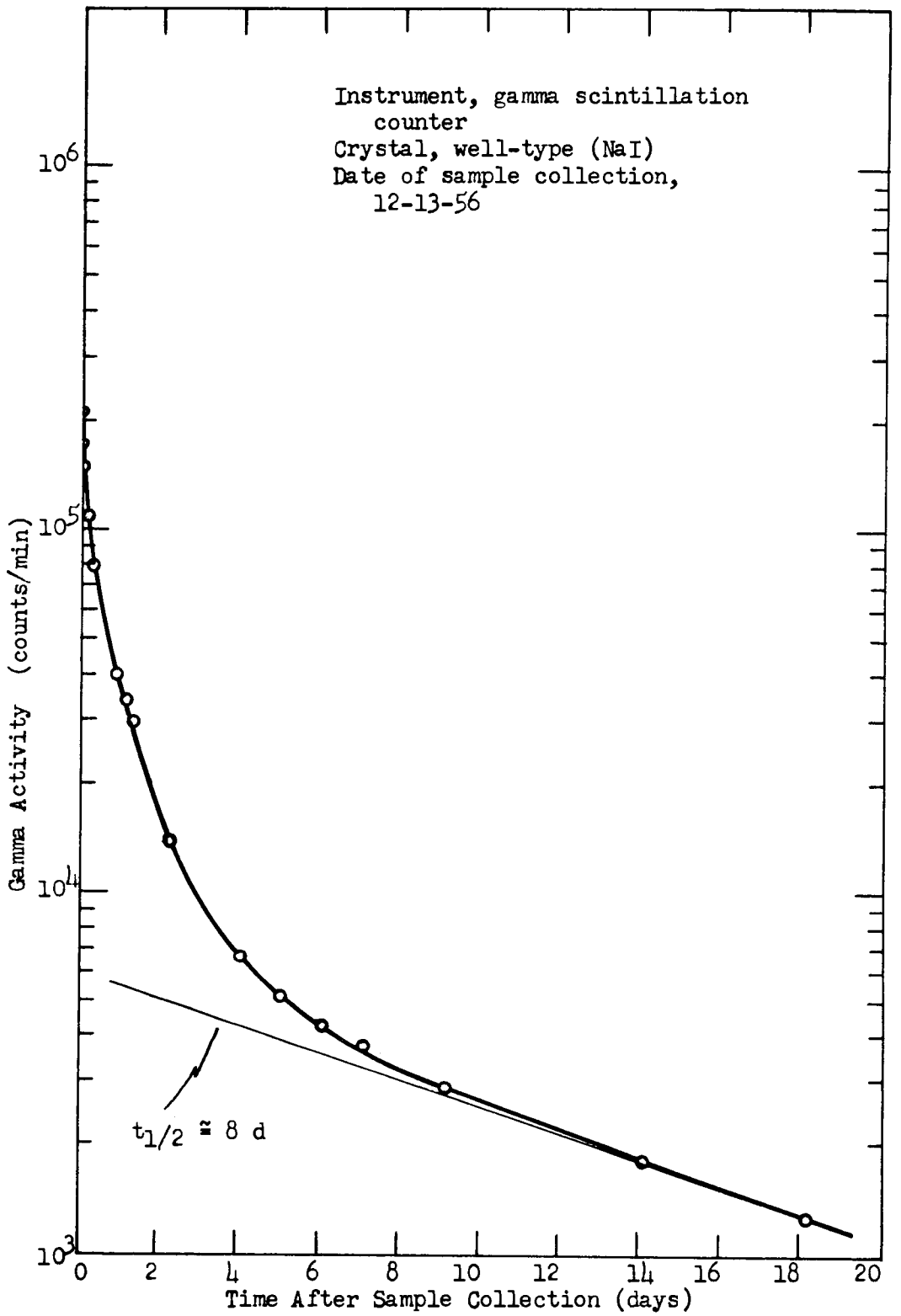

Fig. 18. Gross Gamma Decay for Influent to LITR Cation Exchanger: Silver Group.

The rapidly decaying 0.66 Mev gamma peak in Curve A is indicative of 2.3 h I^{132}. Similarly, the intermediate gross gamma decay shown in Figure 18 also suggests the presence of I^{132}.

Gamma spectral data for the Acid Sulfide Group are shown in Figure 19. The 0.51 and 1.03 Mev peaks indicate the presence of the positron emitter 12.8 h Cu^{64}. This is further substantiated by the presence of the 1.34 Mev gamma peak. The 0.14 Mev peak is indicative of 6.0 h Tc^{99m}. Gross gamma decay data shown in Figure 20 confirm these indications.

Gamma spectral data for the Hydroxide Group are shown in Figure 21. The 0.10 and 0.22 Mev peaks are indicative of 2.33 d Np^{239}. Confirmation is given by the gross gamma decay data as shown in Figure 22 and the subsequent single channel spectrum showing the characteristic 0.09, 0.21 and 0.28 Mev gamma peaks as shown in Figure 23. The 0.53 Mev peak is indicative of 1.87 h F^{18} and 50 m Y^{91m}. The 0.83 Mev peak represents 2.58 h Mn^{56}. The 1.36 Mev peak is possibly indicative of 15.0 h Na^{24}. Yttrium-91m, Mn^{56} and Na^{24} are confirmed in later chemical groups.

Gamma spectral data for the Basic Sulfide Group are shown in Figure 24. The 0.10 and 0.22 Mev peaks again show the presence of 2.33 d Np^{239}. The 0.84 and 1.78 Mev peaks and gross gamma decay data as shown in Figure 25 confirm the presence of 2.58 h Mn^{56}. The long lived 1.16 and 1.34 Mev peaks and the chemistry of this particular sample confirm the presence of 5.2 y Co^{60}.

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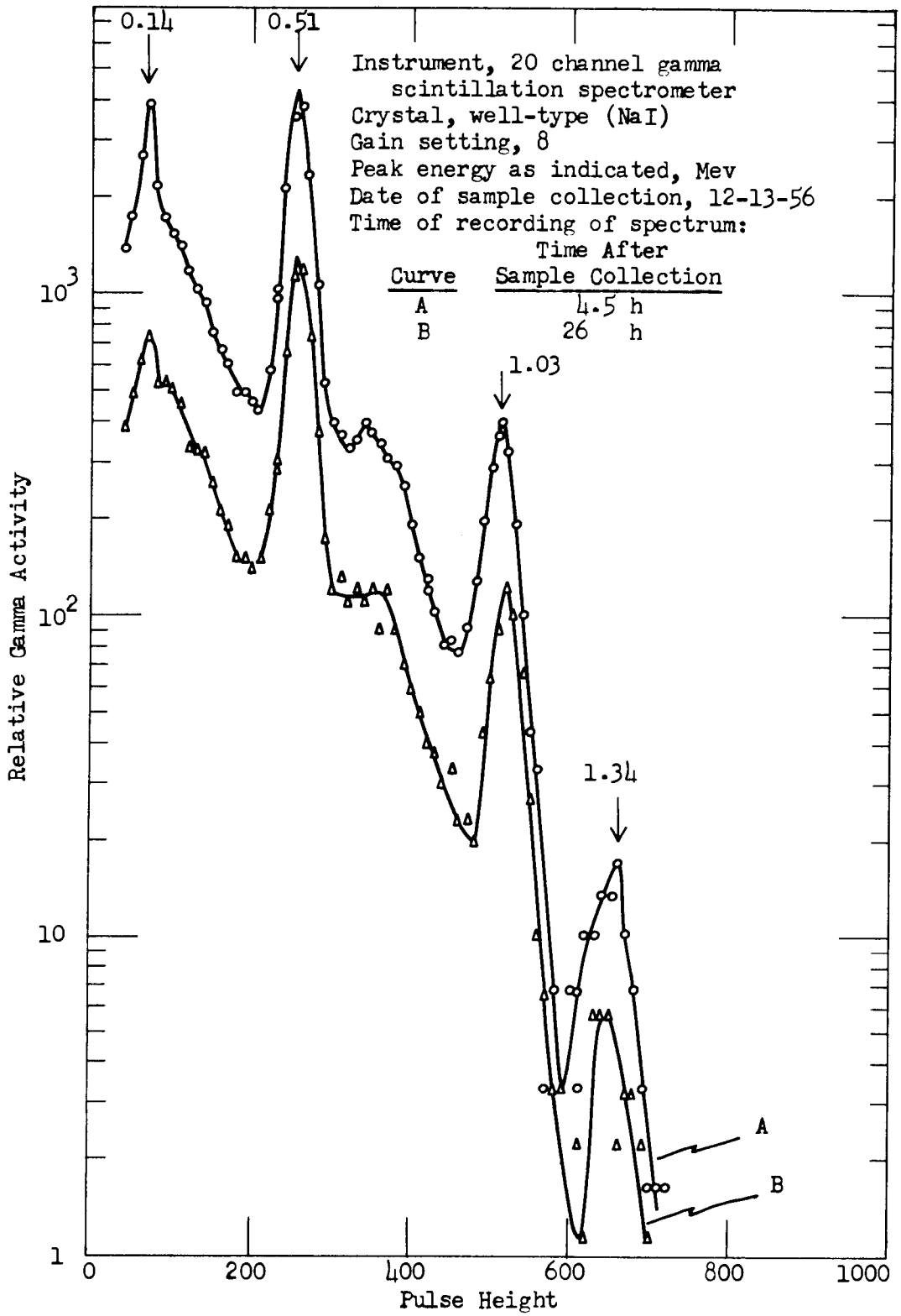

Fig. 19. Gamma Spectral Data for Influent to LITR Cation Exchanger: Acid Sulfide Group.

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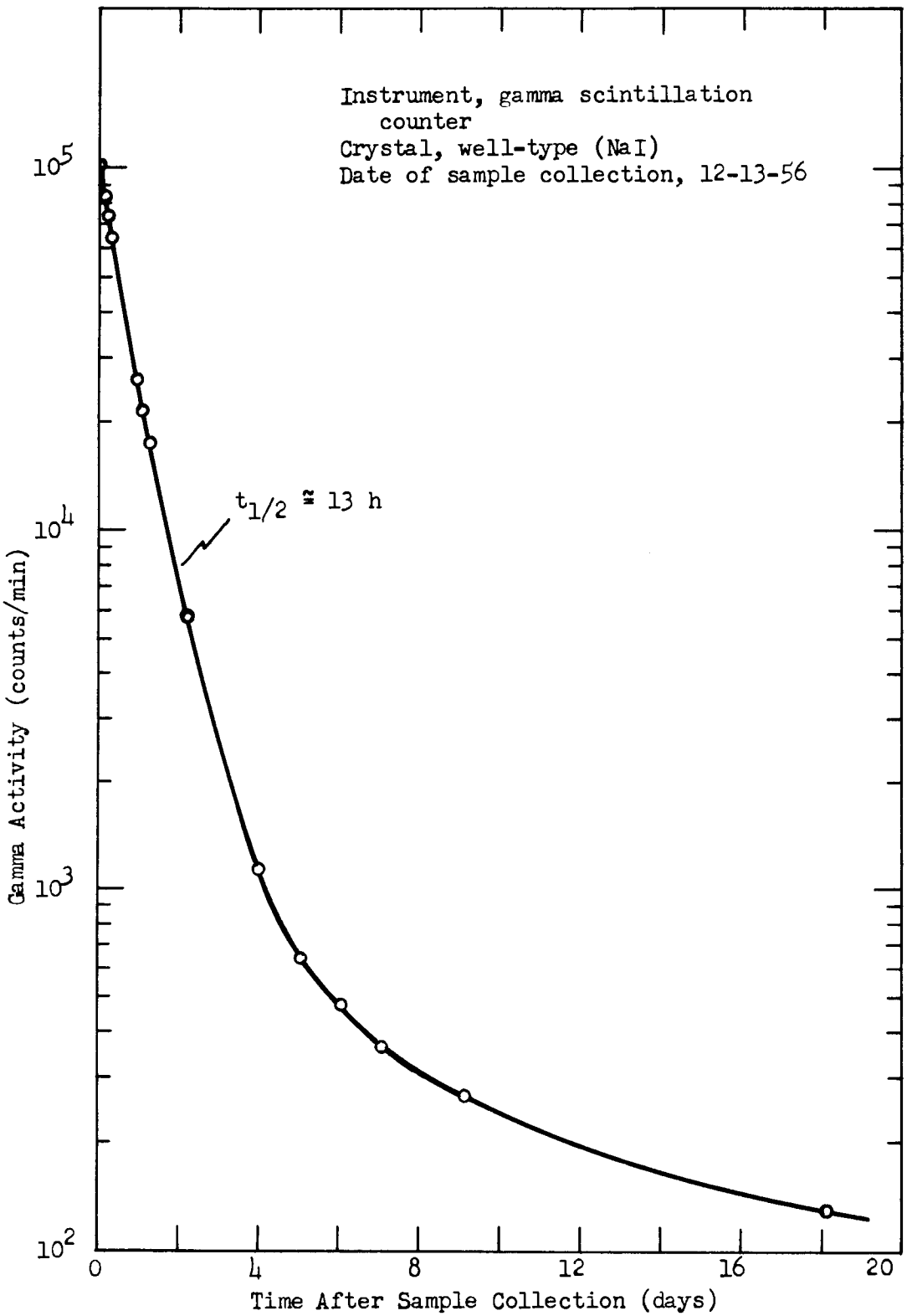

Fig. 20. Gross Gamma Decay for Influent to LITR Cation Exchanger: Acid Sulfide Group.

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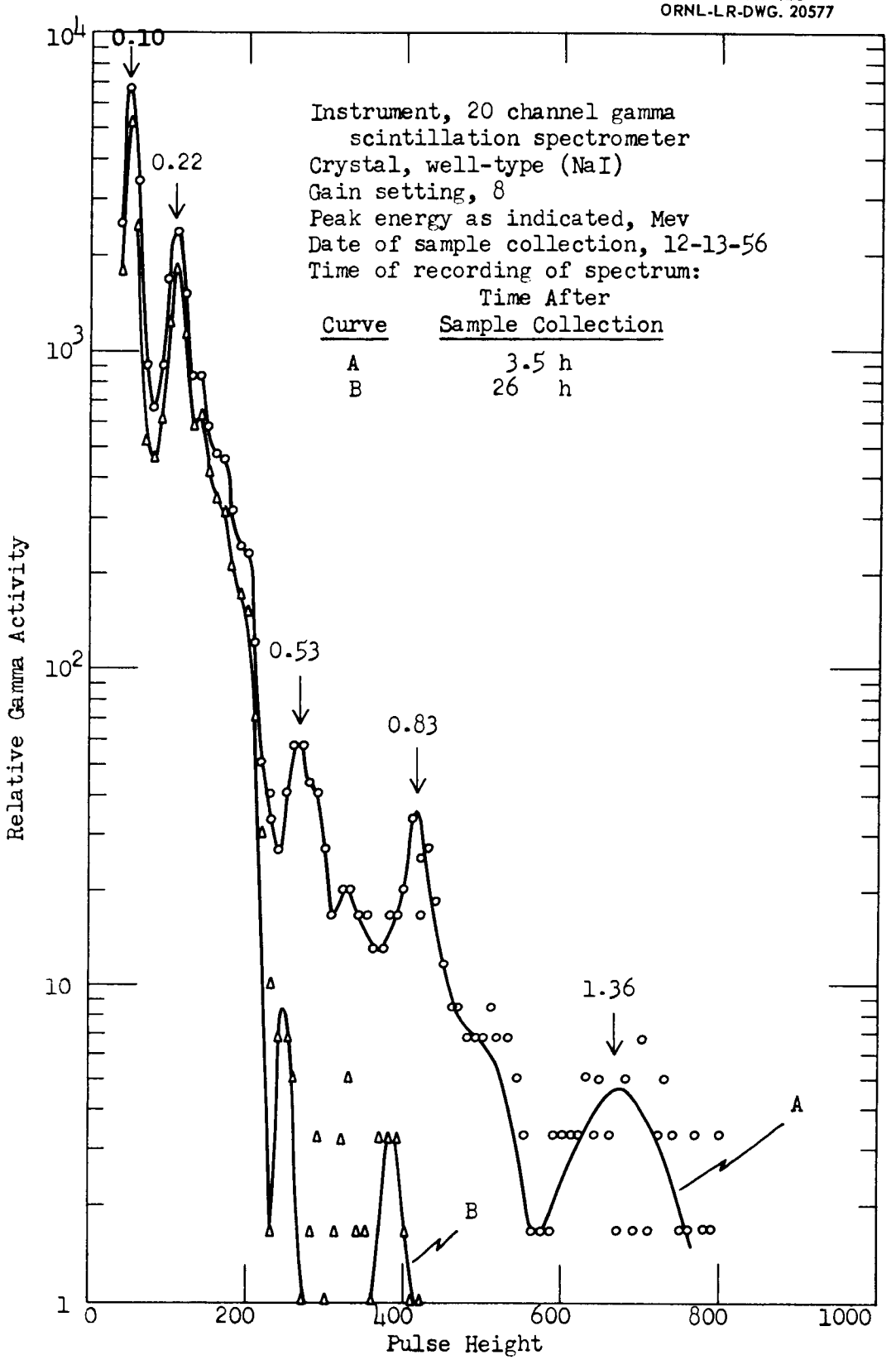

Fig. 21. Gamma Spectral Data for Influent to LITR Cation Exchanger: Hydroxide Group.

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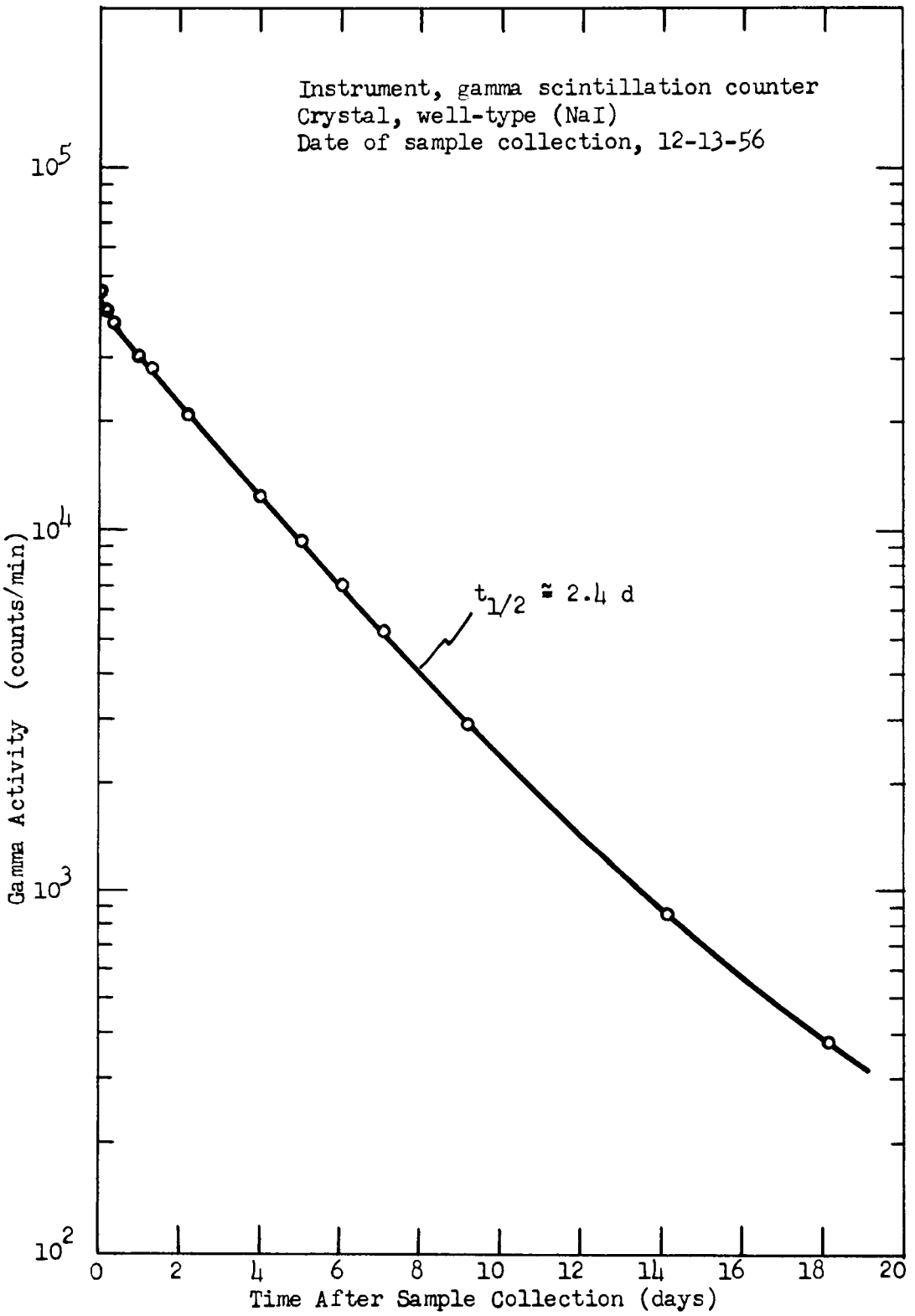

Fig. 22. Gross Gamma Decay for Influent to LITR Cation Exchanger: Hydroxide Group.

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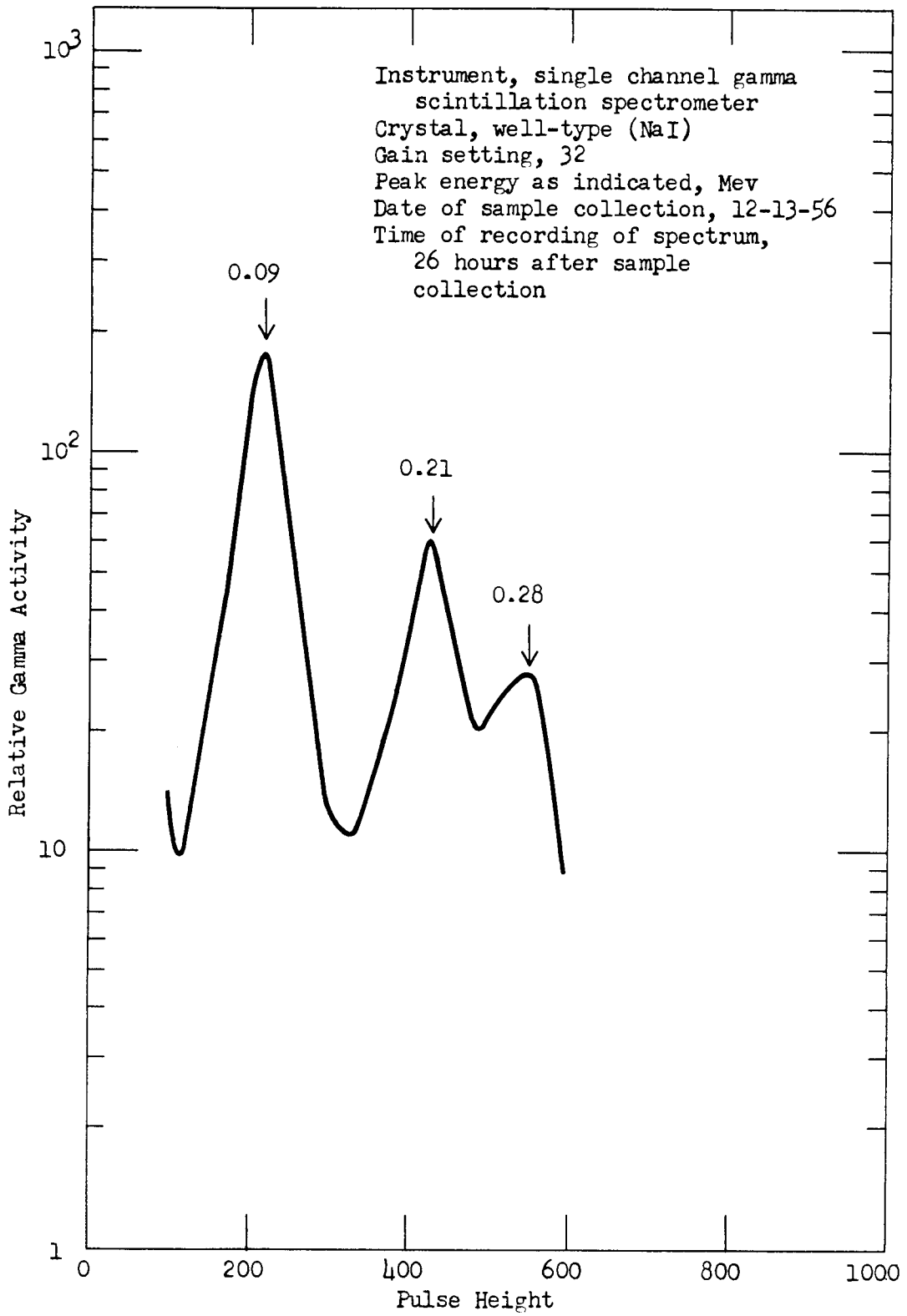

Fig. 23. Gamma Spectral Data for Influent to LITR Cation Exchanger: Hydroxide Group.

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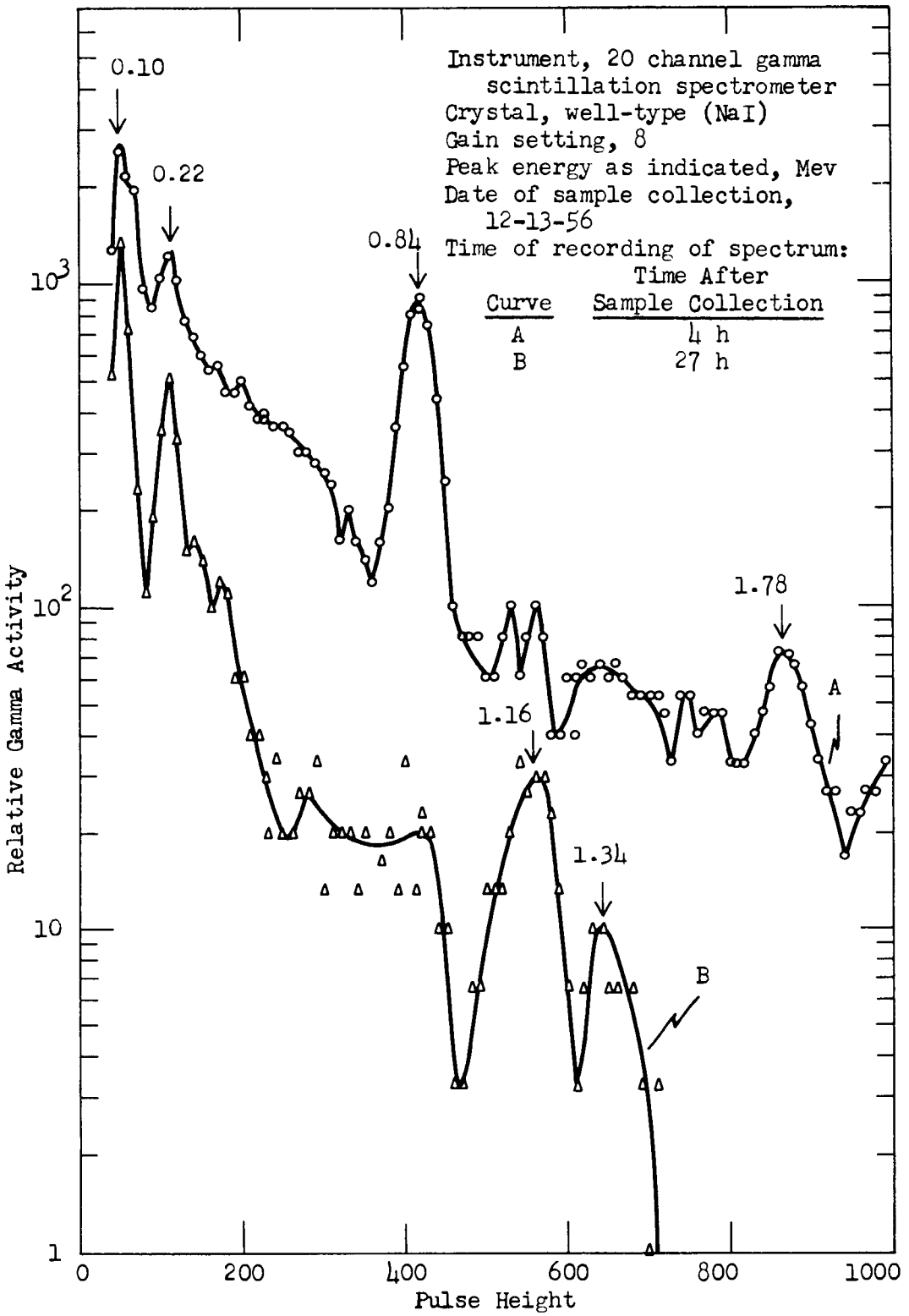

Fig. 24. Gamma Spectral Data for Influent to LITR Cation Exchanger: Basic Sulfide Group.

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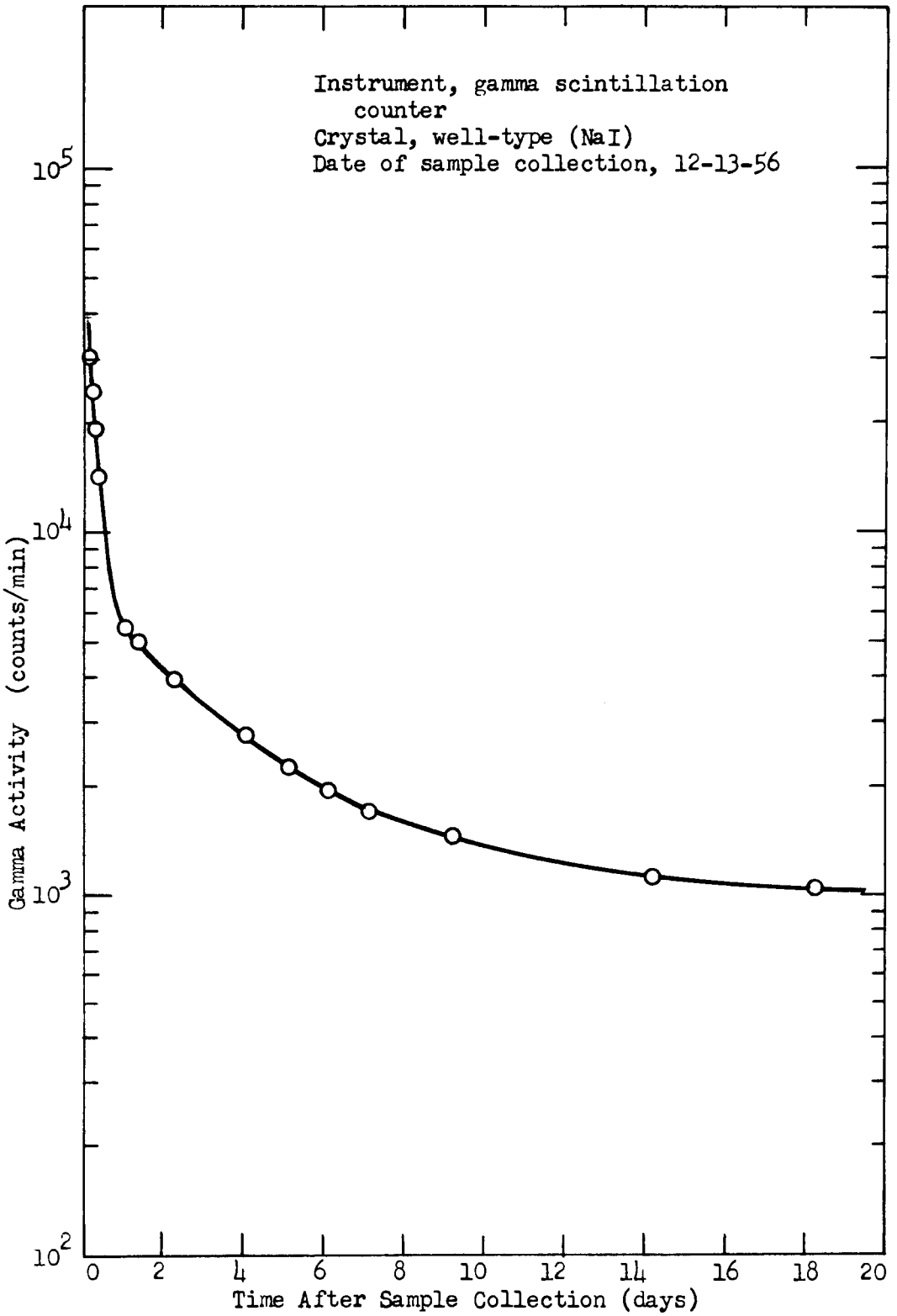

Fig. 25. Gross Gamma Decay for Influent to LITR Cation Exchanger: Basic Sulfide Group.

Gamma spectral data for the Alkaline Earth Group are shown in Figure 26. The 0.10 and 0.22 Mev peaks of Curve A are again indicative of 2.33 d Np^{239}. Half life studies of the 0.56, 0.76 and 1.05 Mev peaks showed them to be associated with 9.7 h Sr^{91} + Y^{91m}. As was mentioned earlier, evidence for 50 m Y^{91m} had been found in the Hydroxide Group. The 1.39 Mev peak is possibly indicative of 2.7 h Sr^{92}. The 0.10 and 0.30 Mev peaks shown in Curve C of the same figure are representative of 27.4 d Pa^{233}. The 0.16 Mev peak is indicative of 12.8 d Ba^{140}. Barium-140 is confirmed by the presence of the 0.47 and 0.79 Mev peaks of its daughter 40.2 h La^{140}. A LaF_3 precipitate of a similar sample also confirmed the presence of La^{140}. Decay data for this group are shown in Figure 27.

Gamma spectral and decay data for the Soluble Group, as shown in Figures 28 and 29, showed the predominant source of activity in this group to be 15.0 h Na^{24}. The 1.34 and 2.75 Mev peaks are representative of this nuclide. The other two peaks shown on the spectral curve are double and single escape peaks resulting from pair production by the higher energy photon (Siegbahn, 1955).

A summary of radioactive nuclides identified in the cation exchanger influent as well as an estimate of their concentrations is given in Table III. Although, as before, the quantitative data indicate orders of magnitude only, being calculated using an assumed average chemical yield of 75% and gamma counting efficiency of 33%, specific quantitative determinations of several nuclides in similar samples compared favorably with the estimates reported here.

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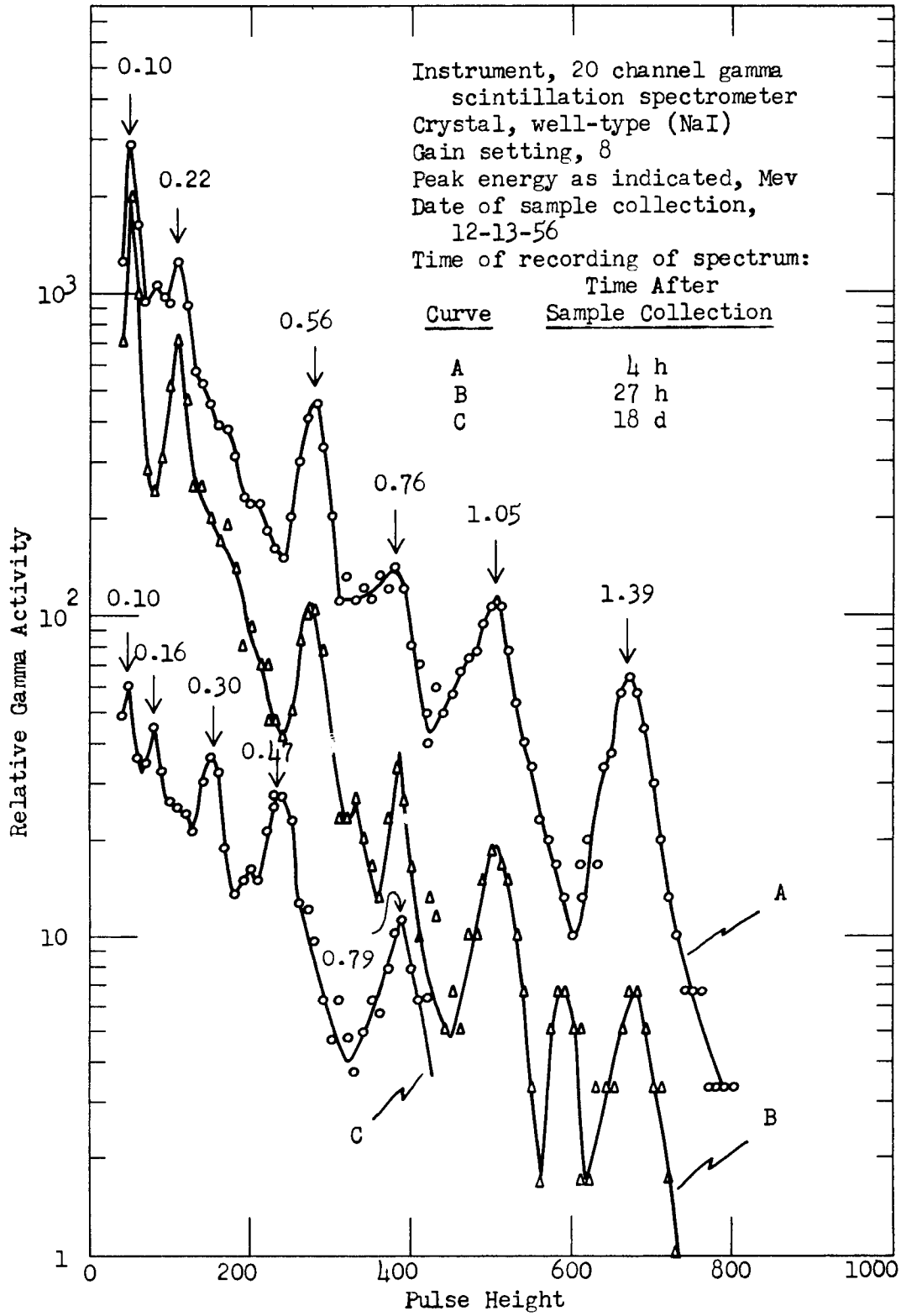

Fig. 26. Gamma Spectral Data for Influent to LITR Cation Exchanger: Alkaline Earth Group.

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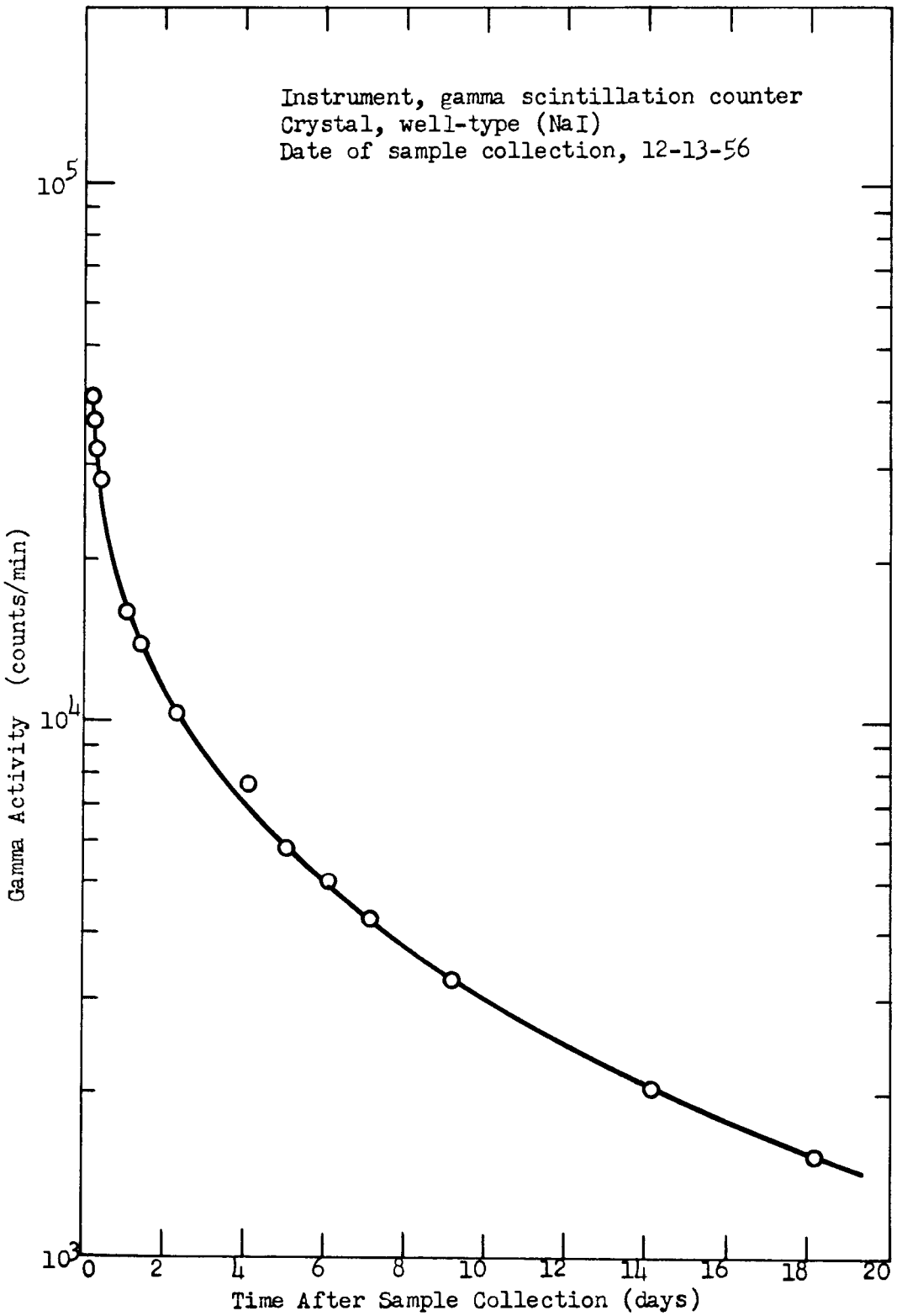

Fig. 27. Gross Gamma Decay for Influent to LITR Cation Exchanger: Alkaline Earth Group.

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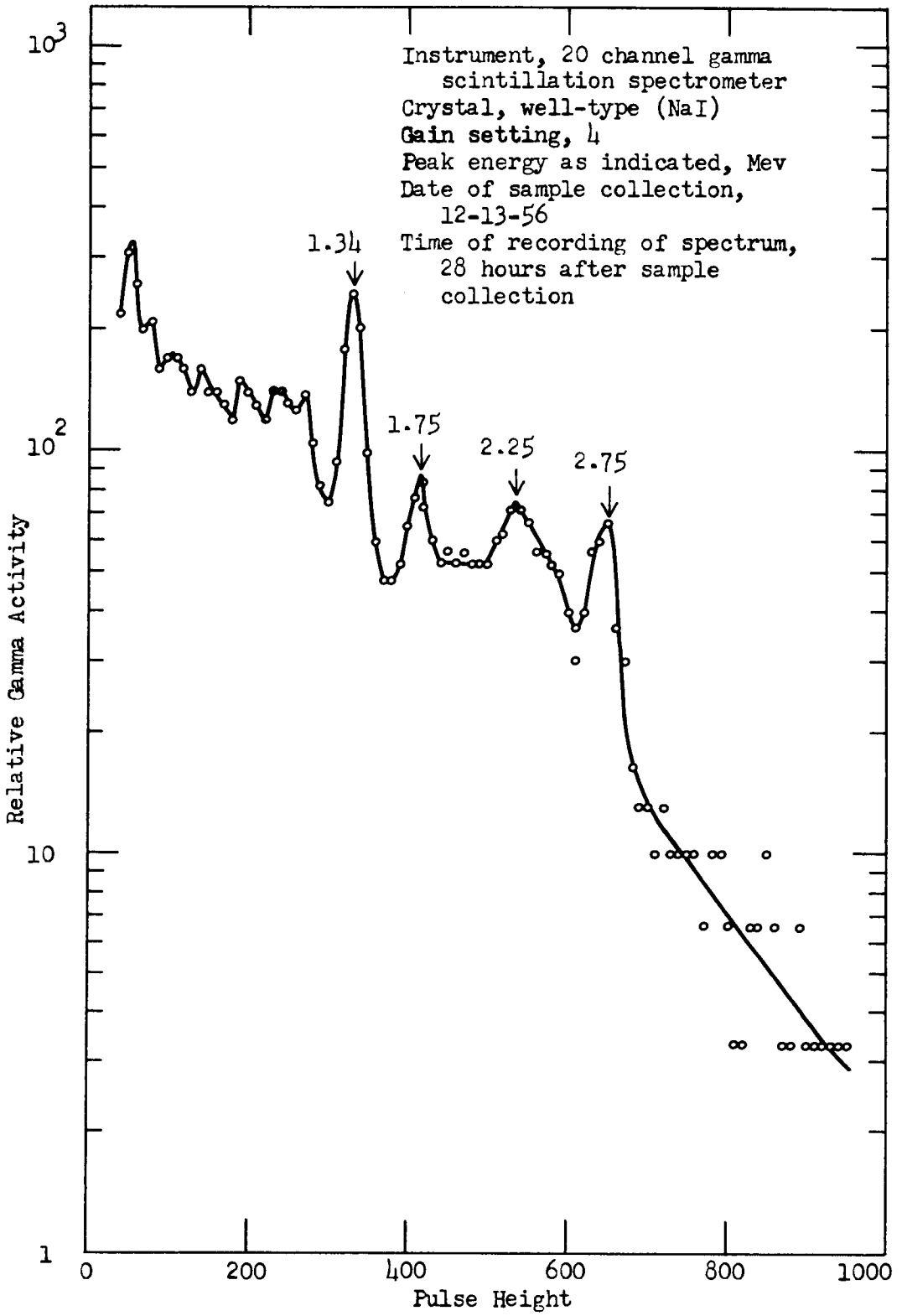

Fig. 28. Gamma Spectral Data for Influent to LITR Cation Exchanger: Soluble Group.

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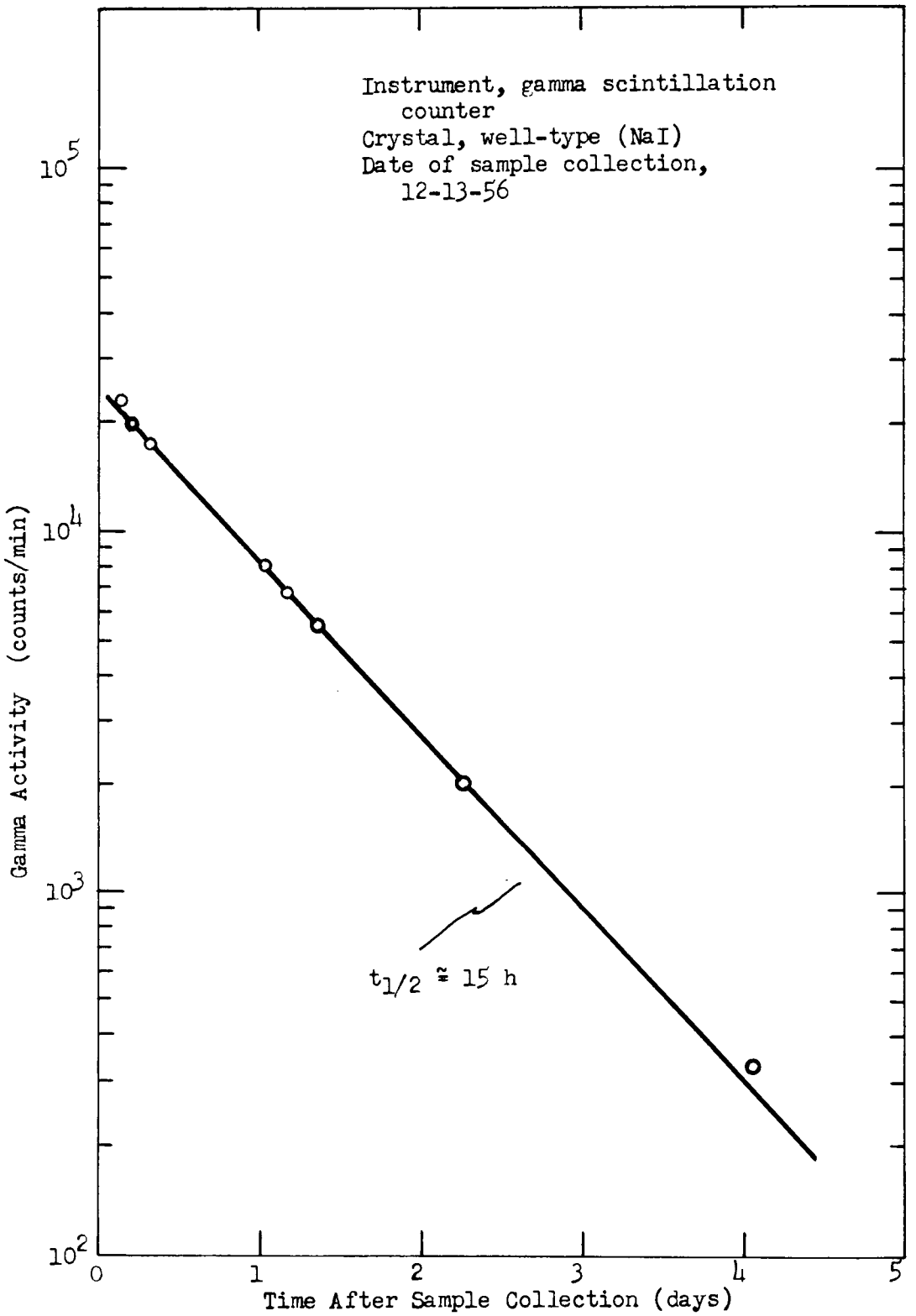

Fig. 29. Gross Gamma Decay for Influent to LITR Cation Exchanger: Soluble Group.

TABLE III

Radioactive Nuclides Identified in the Influent to the LITR Cation Exchanger

Date of sample collection, 12-13-56

Chemical Group	Radioactive Nuclide	Half Life	Estimated Concentration (μc/ml)*
Silver	I^{131}	8.05 d	8×10^{-5}
	+ Xe^{131m_1}	4.8×10^{-10} s	
	I^{132}	2.3 h	
	I^{133}	21 h	6×10^{-4} } 3×10^{-3}
	+ Xe^{133}	5.27 d	
	+ Cs^{133m}	6.0×10^{-9} s	
	I^{135}	6.7 h	
	+ Xe^{135}	9.2 h	
	+ Cs^{135m}	2.8×10^{-10} s	
Acid Sulfide	Cu^{64}	12.8 h	1×10^{-3}
	Tc^{99m}	6.0 h	7×10^{-4}
Hydroxide	Np^{239}	2.33 d	5×10^{-4}
	Mn^{56}	2.58 h	
	Na^{24}	15.0 h	
	Y^{91m}	50 m	1×10^{-4}
Basic Sulfide	Np^{239}	2.33 d	1×10^{-4}
	Mn^{56}	2.58 h	7×10^{-4}
	Co^{60}	5.2 y	1×10^{-5}
Alkaline Earth	Np^{239}	2.33 d	2×10^{-4}
	Sr^{91}	9.7 h	5×10^{-4}
	+ Y^{91m}	50 m	
	Pa^{233}	27.4 d	1×10^{-5}
	Ba^{140}	12.8 d	1×10^{-5}
	+ La^{140}	40.2 h	
Soluble	Na^{24}	15.0 h	3×10^{-3}

* At time of sample collection.

C. Cation Exchanger Effluent

The second cooling water sample was collected at the effluent from the cation exchanger. Identification of the radioactive nuclides remaining in the cooling water after passage through the cation exchanger was necessary for several reasons: (a) it would reveal information as to the ionic behavior of the radioactive nuclides in the water, (b) it would aid in the evaluation of cation exchangers in cooling water purification systems.

The sample was chemically separated as previously outlined. Because of the lower over-all activity expected, a sample volume of 280 milliliters was taken as opposed to the 140 milliliter sample taken for the cation exchanger influent analysis.

Again, for spectral studies the primary instrument used was the 20 channel gamma scintillation spectrometer, the single channel unit serving only for supplementary purposes.

Gamma spectral and decay data for the Silver Group are shown in Figures 30 and 31. The similarity between these decay and spectral data and those for the Silver Group of the cation exchanger influent is readily apparent. Again, 8.05 d I^{131}, 2.3 h I^{132}, 21 h I^{133}, 6.7 h I^{135} and their daughter products were concluded to be present in the sample.

Gamma spectral and decay data for the Acid Sulfide Group are shown in Figures 32 and 33. Two prominent gamma peaks are in evidence, namely one at 0.14 Mev and one at 0.57 Mev. A study of the gross gamma decay data and the chemistry of this group reveal

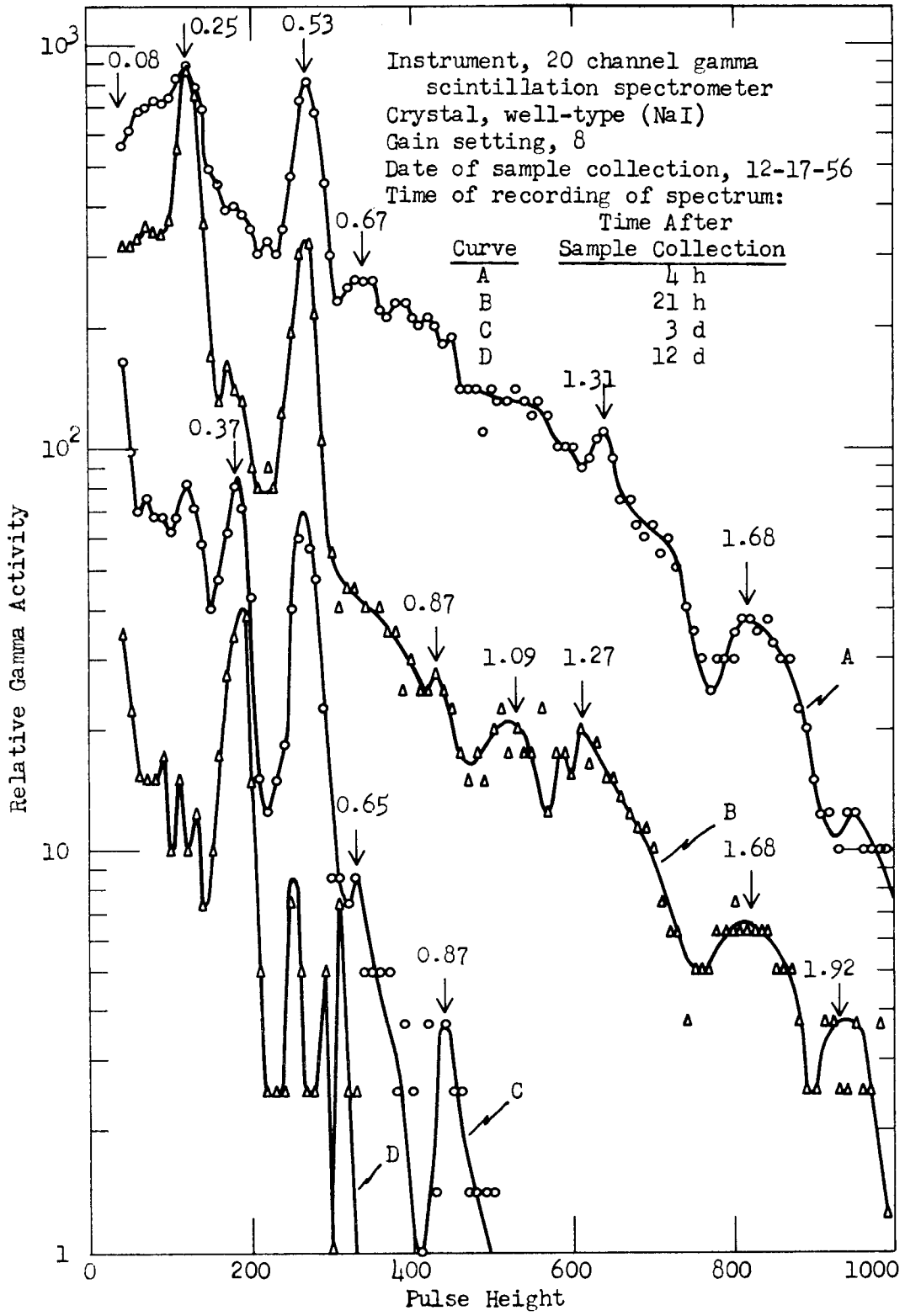

Fig. 30. Gamma Spectral Data for Effluent from LITR Cation Exchanger: Silver Group.

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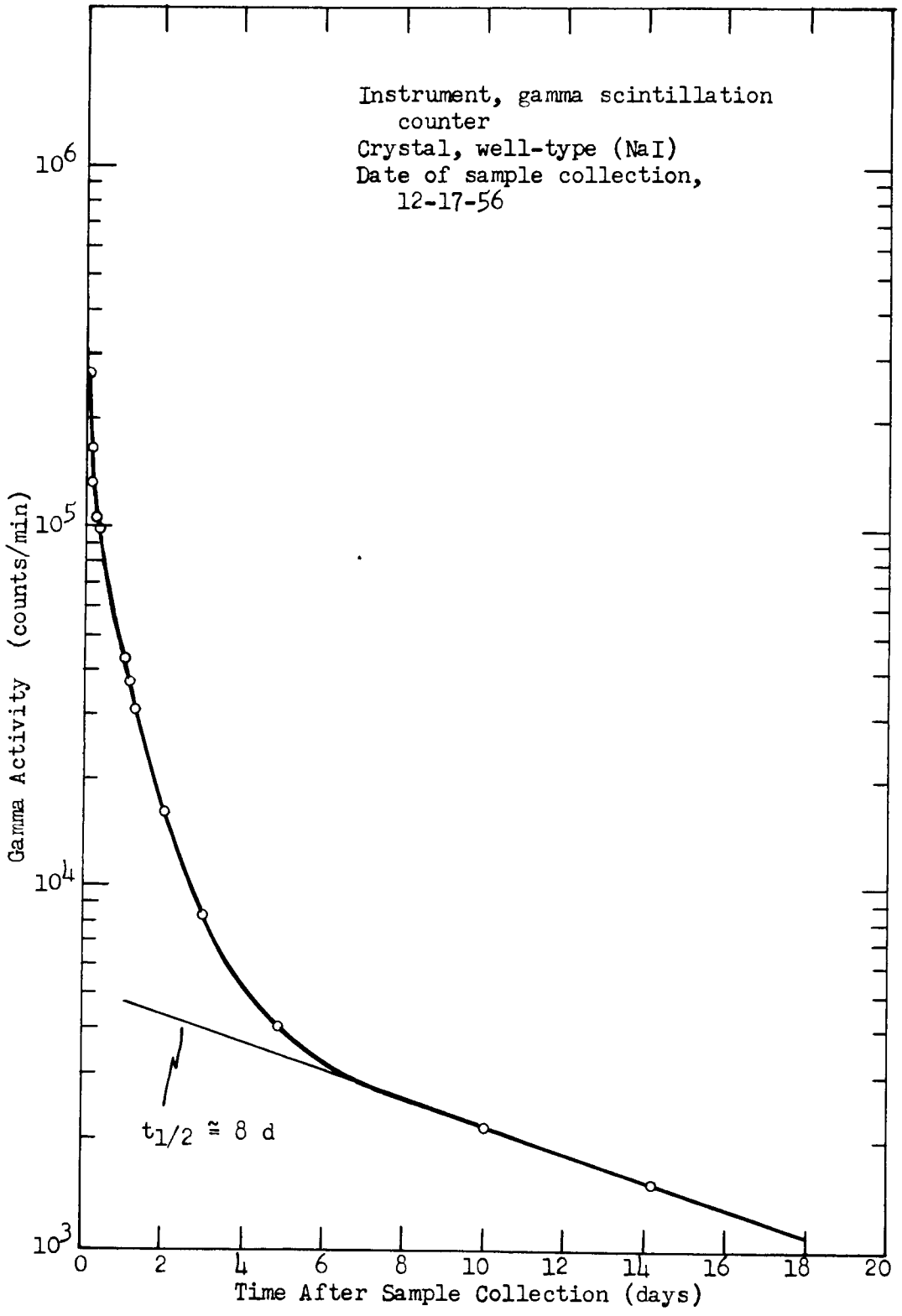

Fig. 31. Gross Gamma Decay for Effluent from LITR Cation Exchanger: Silver Group.

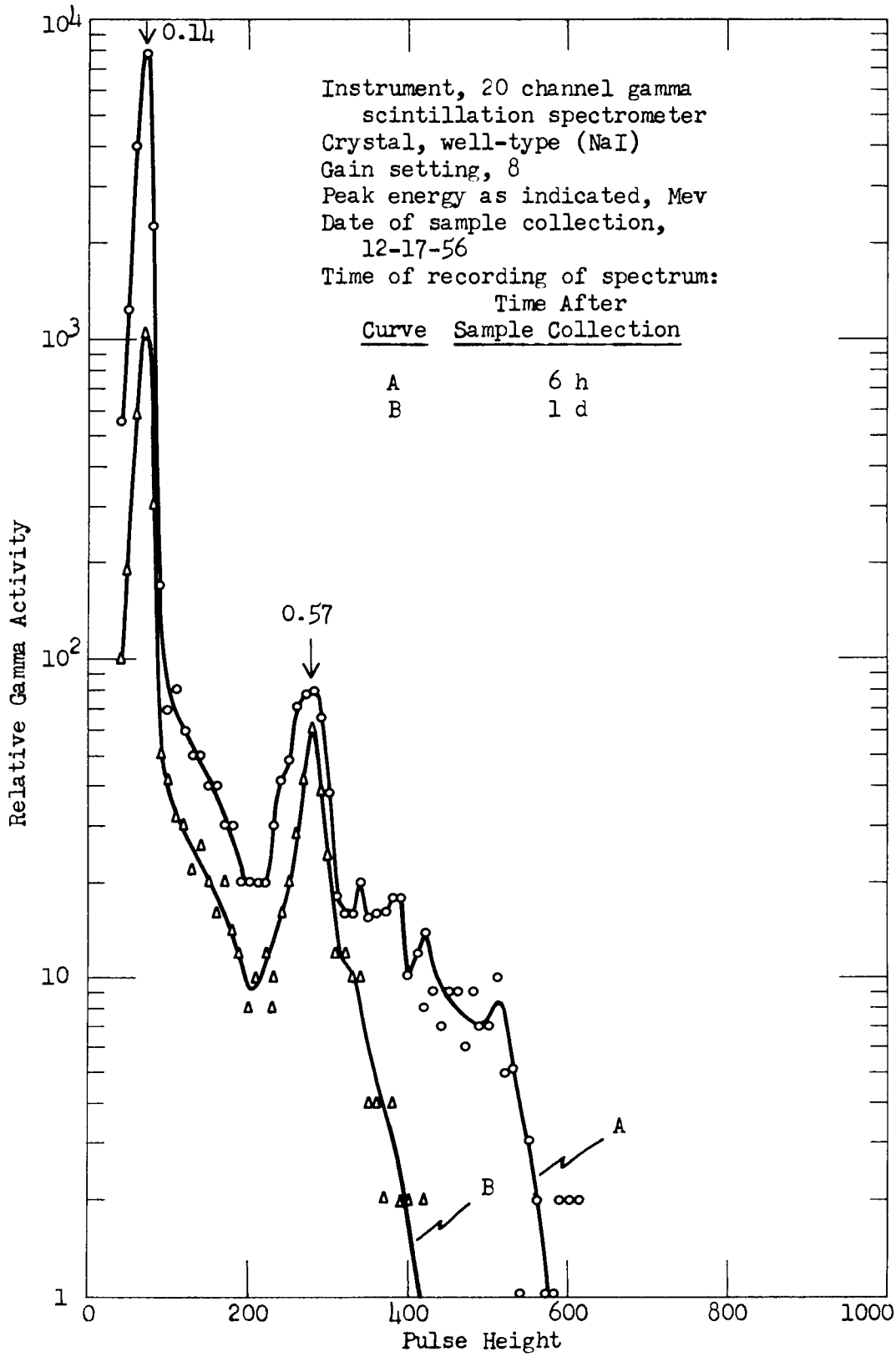

Fig. 32. Gamma Spectral Data for Effluent from LITR Cation Exchanger: Acid Sulfide Group.

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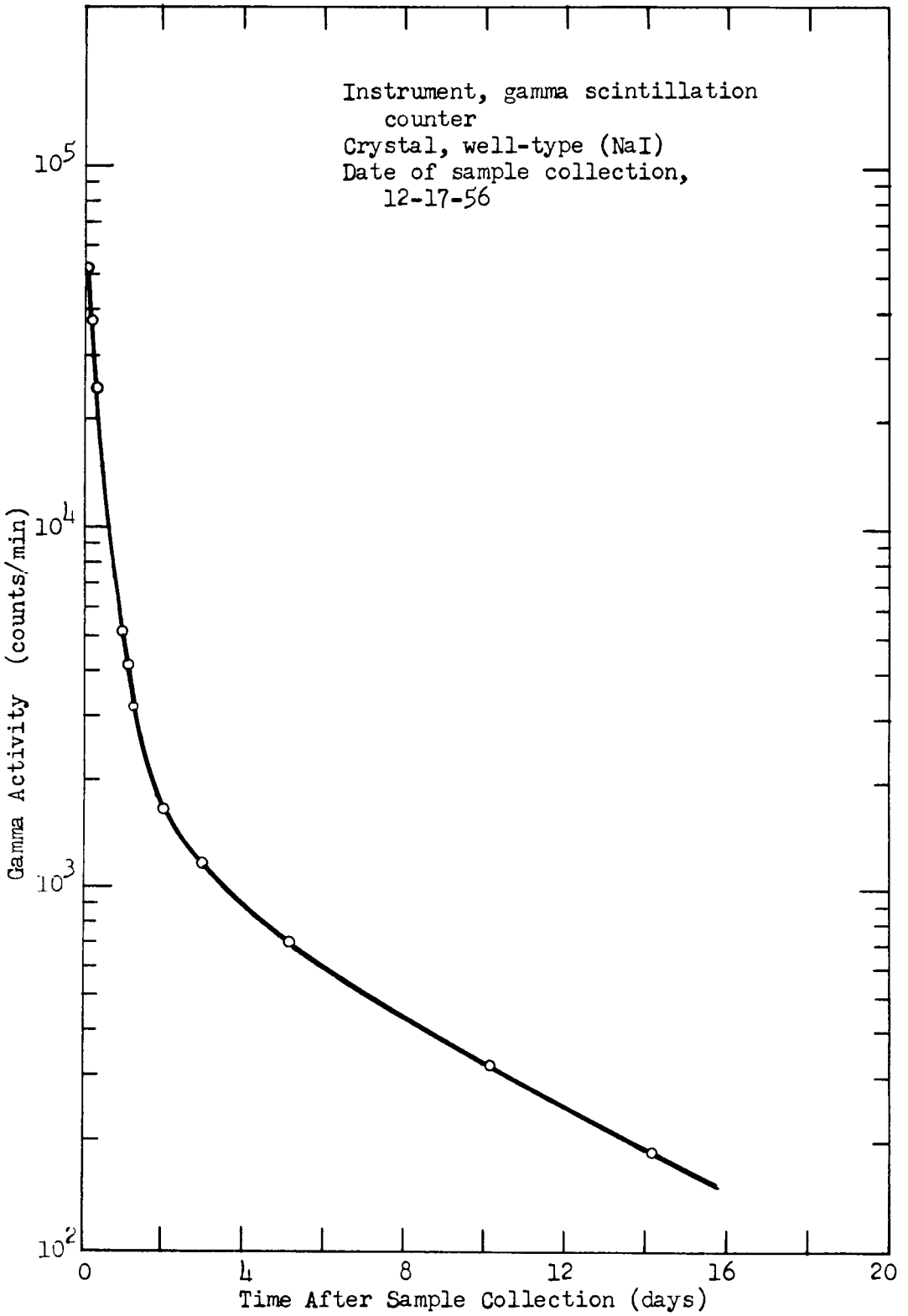

Fig. 33. Gross Gamma Decay for Effluent from LITR Cation Exchanger: Acid Sulfide Group.

these to represent 6.0 h Tc^{99m} and 2.8 d Sb^{122}. Inasmuch as the 0.14 Mev peak decayed with a half life of six hours, it was concluded that little of the longer-lived Mo^{99} parent of Tc^{99m} was present.

Gamma spectral and decay data for the Hydroxide Group are shown in Figures 34 and 35. The 0.14 Mev peak in Curve A is again indicative of 6.0 h Tc^{99m}. The fact that the 0.14 peak was still noticeable after the sample had decayed for 10 days suggests the presence of either 32 d Ce^{141} or the 77 h Mo^{99} parent of Tc^{99m}. The 0.51 and 1.02 Mev peaks decayed with a half life of approximately two hours. It was concluded that they represented the positron emitter 1.87 h F^{18}.

The prominent 0.10 and 0.30 Mev peaks shown in Curve C are indicative of 27.4 d Pa^{233}. Its presence was later confirmed in the Alkaline Earth Group. The 0.23 and 0.67 Mev peaks were not identified.

Gamma spectral and decay data for the Basic Sulfide Group are shown in Figures 36 and 37. Again the presence of the 0.14 Mev peak decaying with a half life of approximately 6 hours indicates the presence of Tc^{99m}. The gross gamma decay curve indicates Tc^{99m} constitutes the major portion of this group.

Gamma spectral and decay data for the Alkaline Earth Group are shown in Figures 38 and 39. Again, the 0.14 Mev peak, as well as the rapid initial decay of the gross gamma curve, indicates the presence of Tc^{99m}. After a decay period of fourteen days, the predominant remaining activity is Pa^{233}, as exemplified by the major peaks at 0.10 and 0.31 Mev coupled with its characteristic smaller peaks at approximately 0.18 and 0.40 Mev (Moore and Reynolds, 1957).

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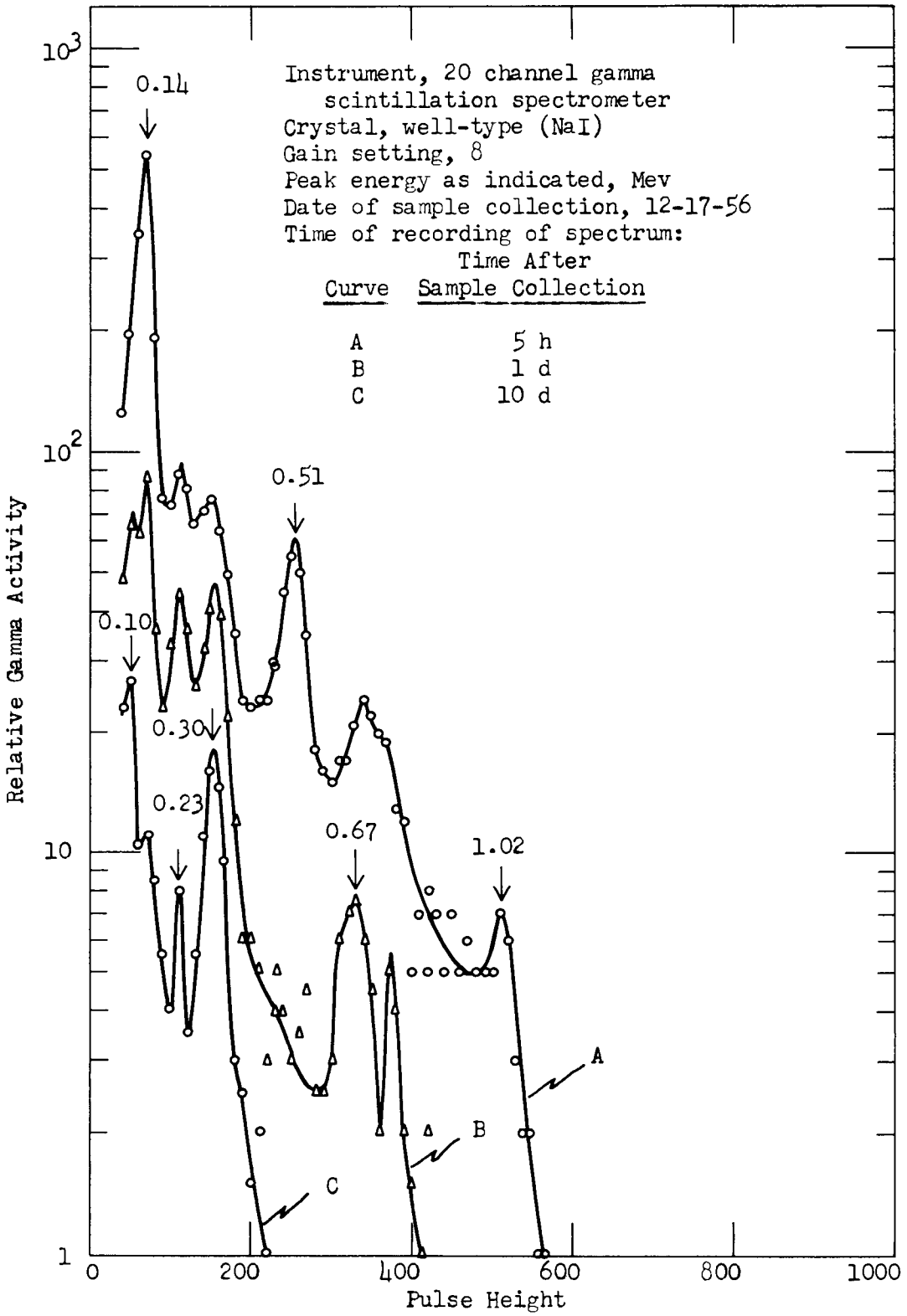

Fig. 34. Gamma Spectral Data for Effluent from LITR Cation Exchanger: Hydroxide Group.

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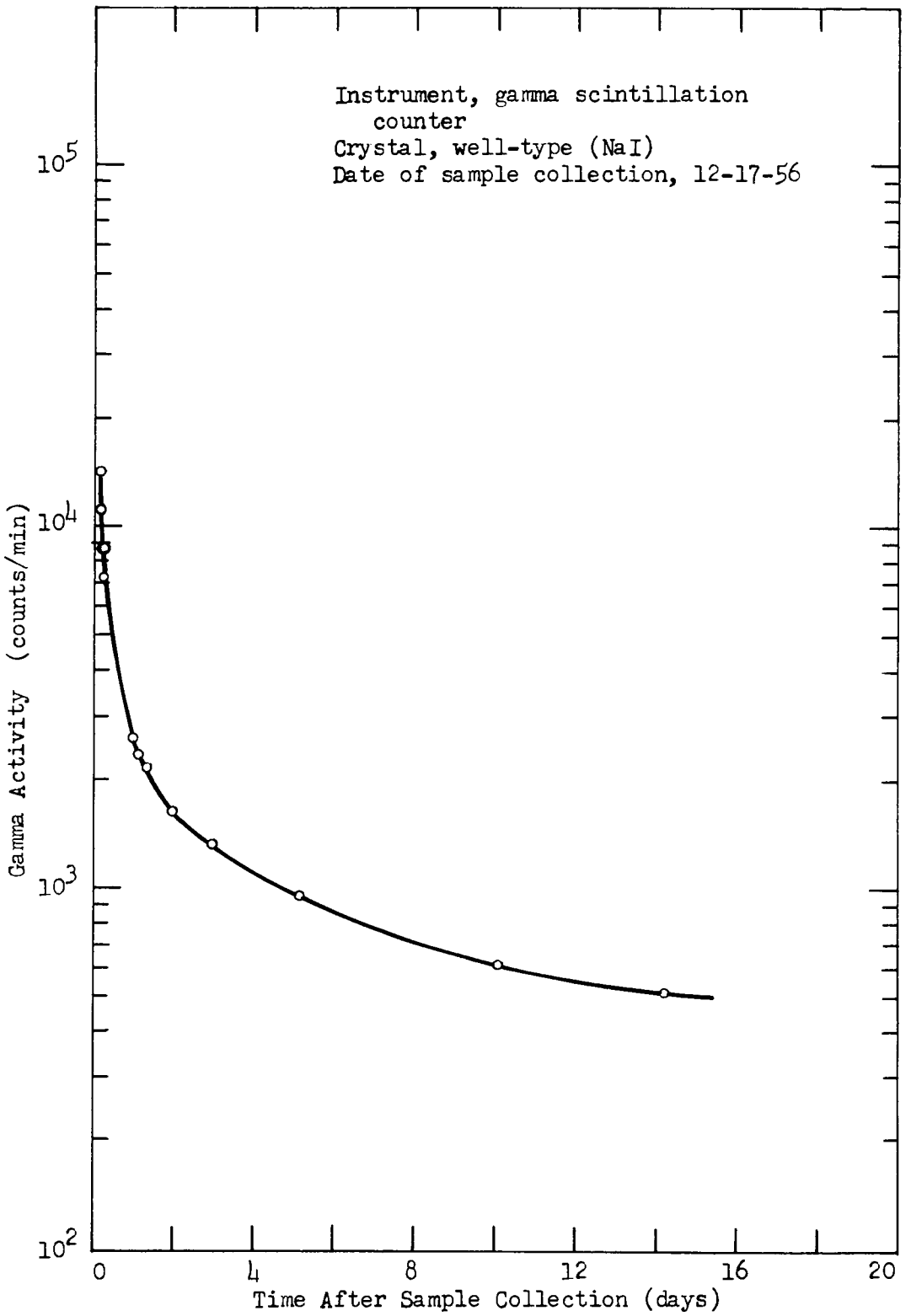

Fig. 35. Gross Gamma Decay for Effluent from LITR Cation Exchanger: Hydroxide Group.

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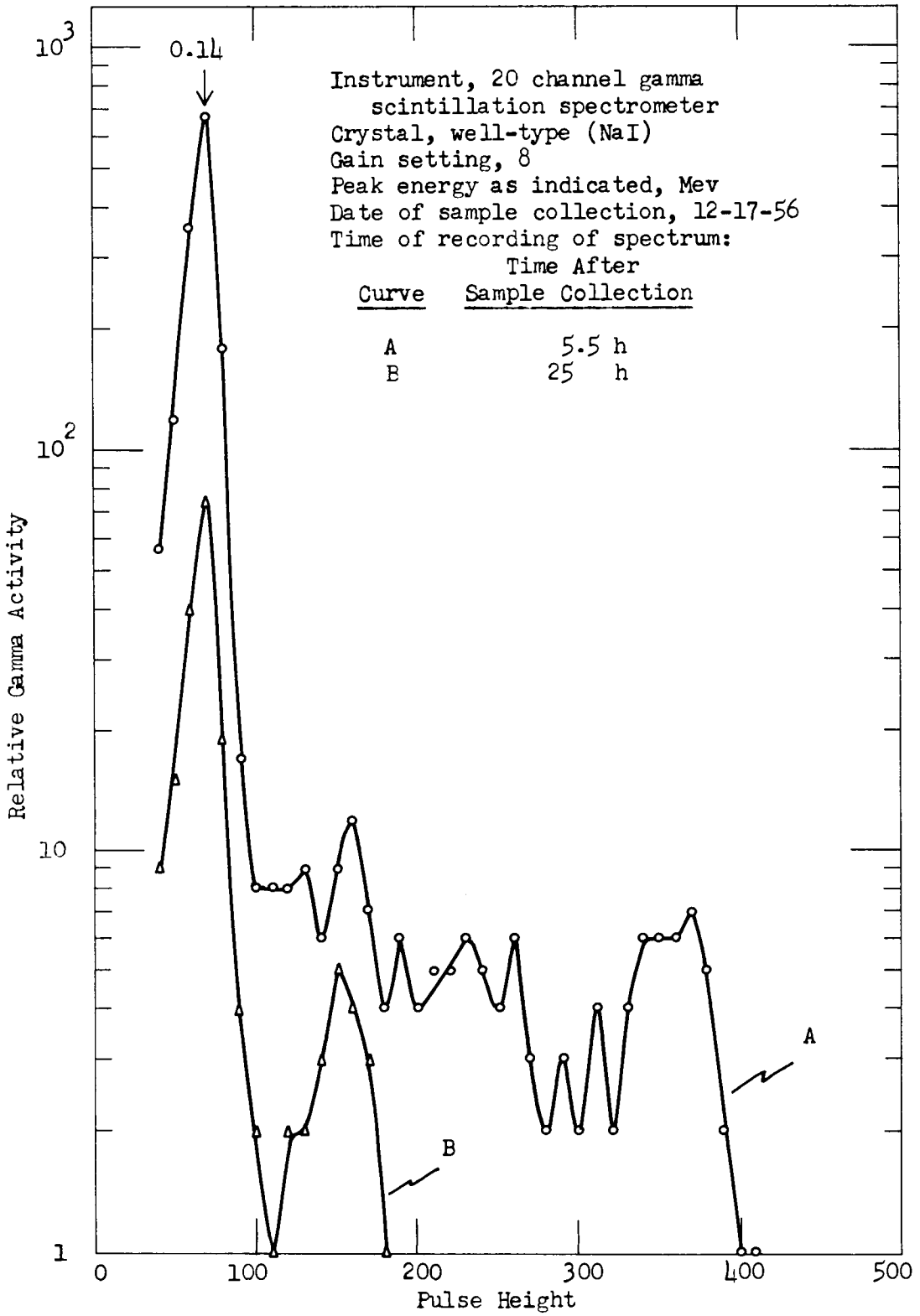

Fig. 36. Gamma Spectral Data for Effluent from LITR Cation Exchanger: Basic Sulfide Group.

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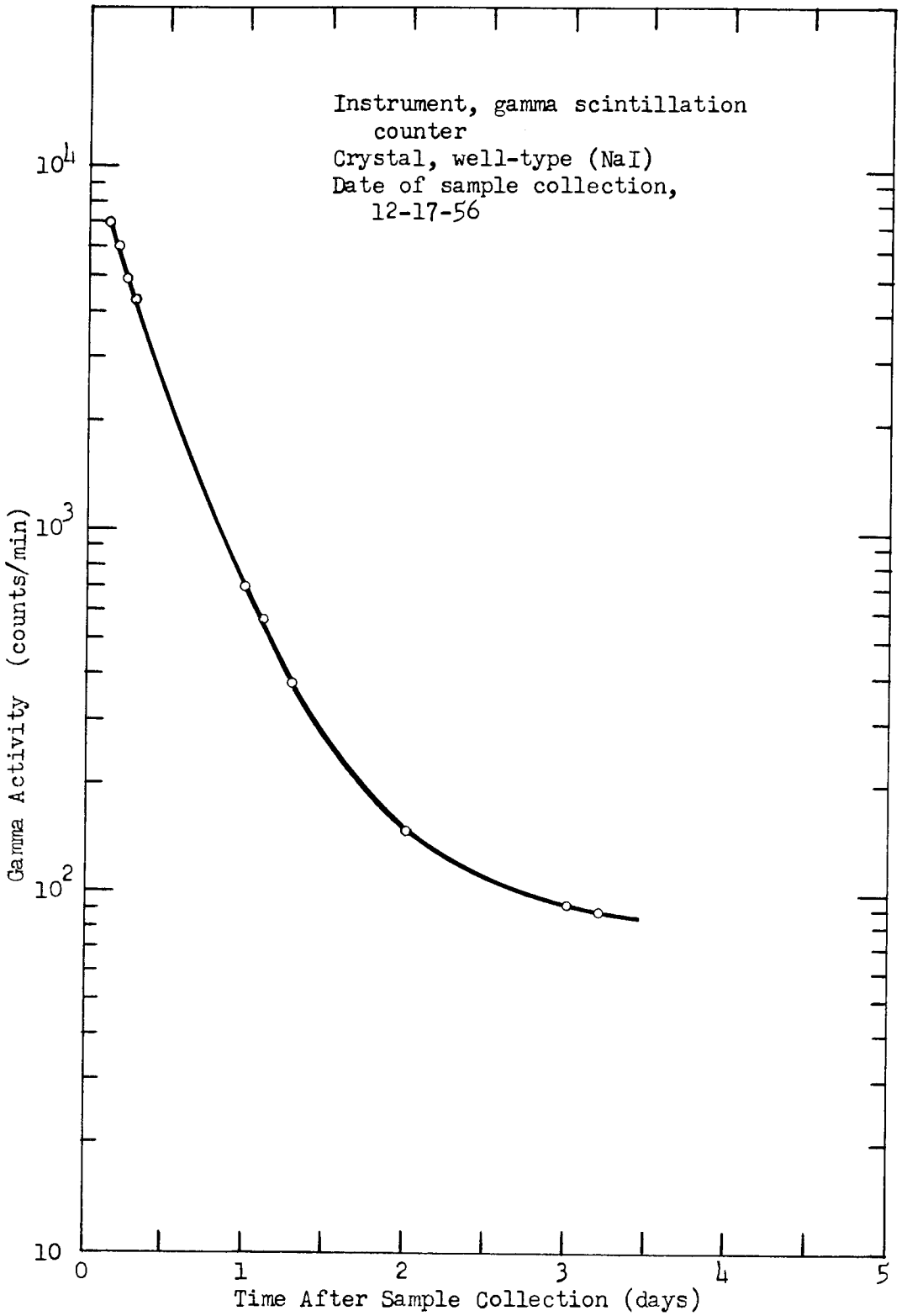

Fig. 37. Gross Gamma Decay for Effluent from LITR Cation Exchanger: Basic Sulfide Group.

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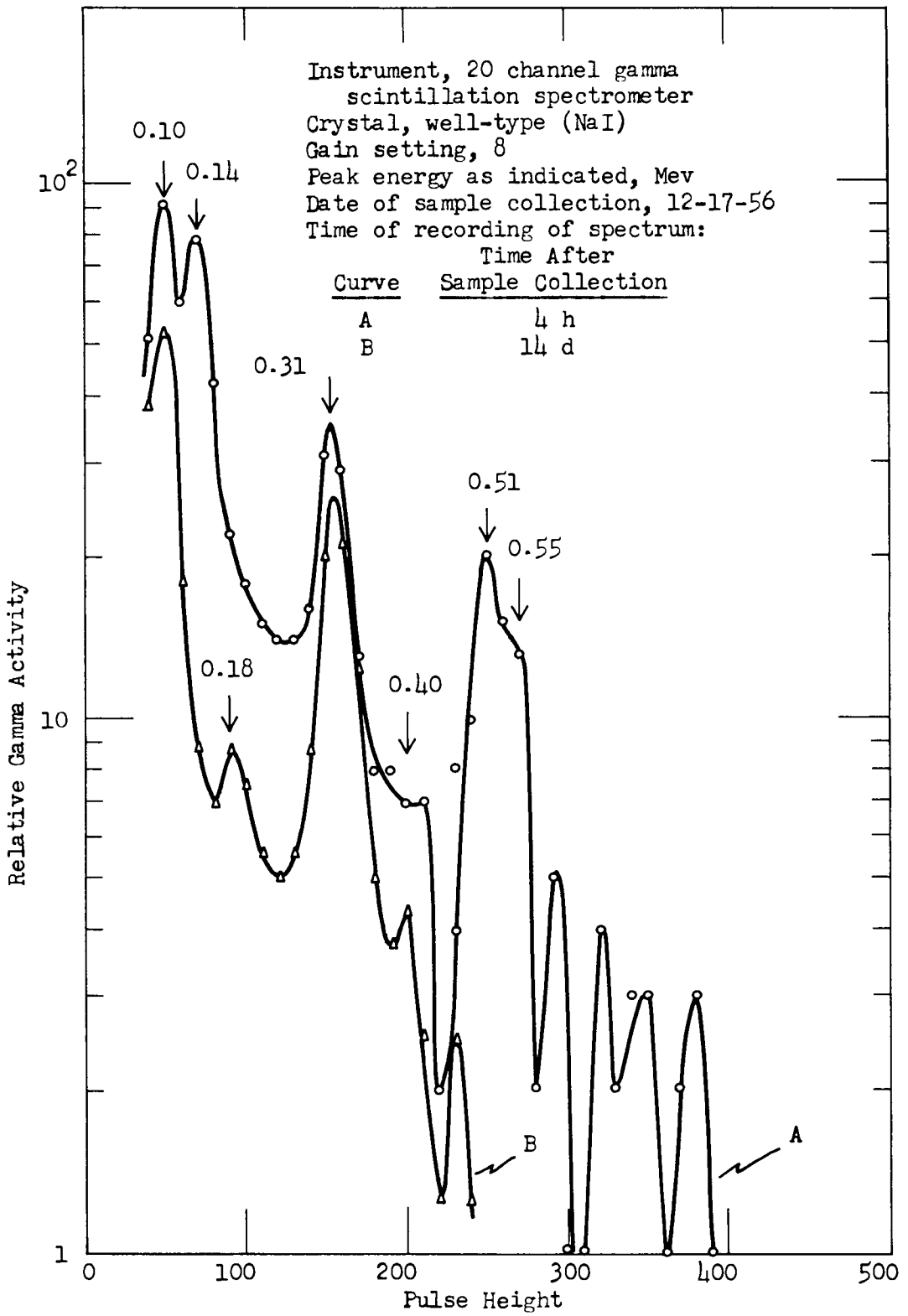

Fig. 38. Gamma Spectral Data for Effluent from LITR Cation Exchanger: Alkaline Earth Group.

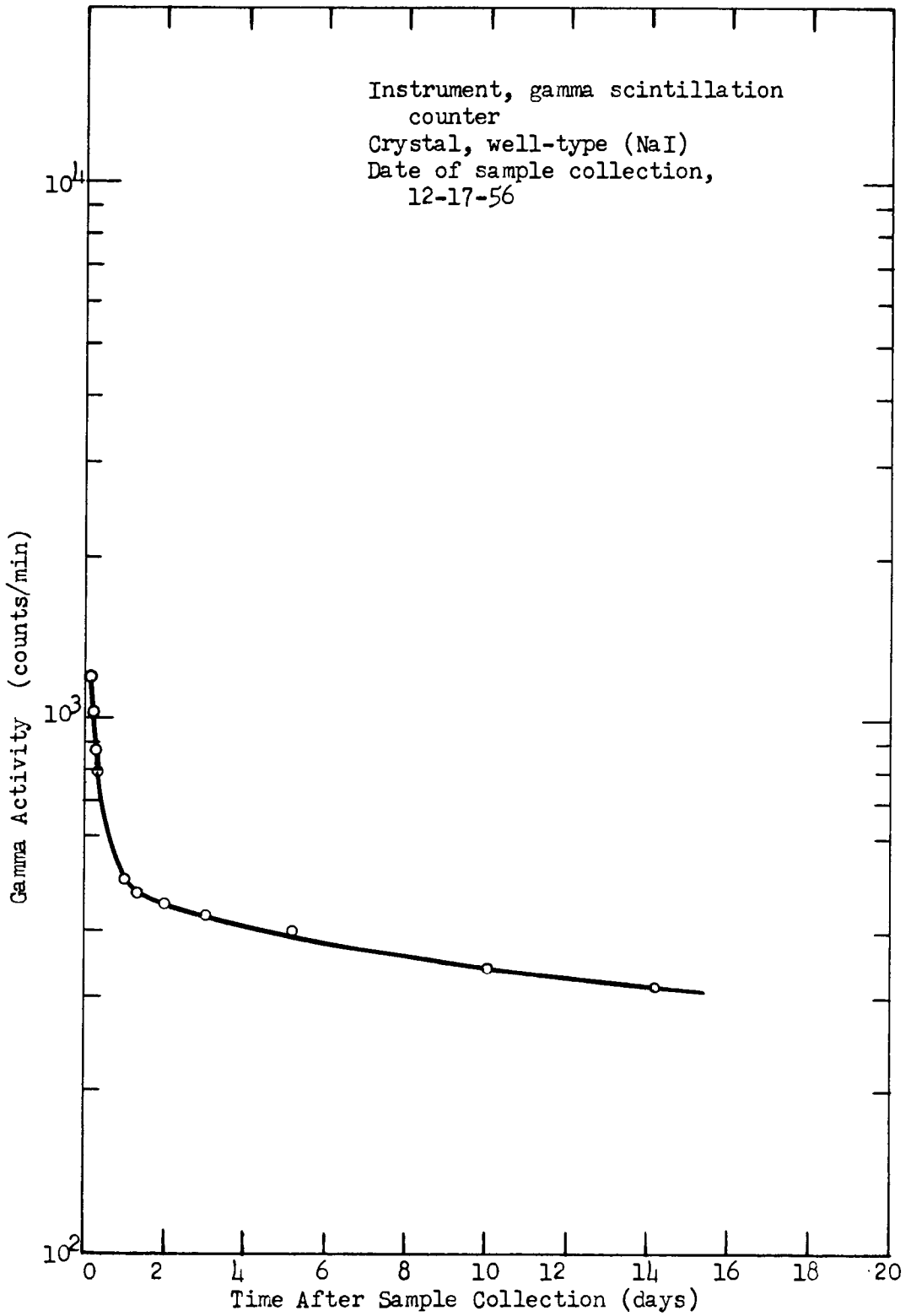

Fig. 39. Gross Gamma Decay for Effluent from LITR Cation Exchanger: Alkaline Earth Group.

The short-lived 0.51 Mev peak again represents 1.87 h F^{18}. The presence of the shoulder on the 0.51 Mev peak perhaps indicates that some Sr^{91} + Y^{91m} remains in the cooling water.

The Soluble Group contained an insignificant amount of radioactive material.

A summary of radioactive nuclides identified in the cation exchanger effluent and their relative concentrations is given in Table IV. As before, the quantitative data are meant to illustrate orders of magnitude only.

D. Anion Exchanger Effluent

The first stage of the cooling water purification system consists of the cation exchanger. Water leaving this unit then passes through an anion exchanger. The effluent from the anion unit was therefore chosen as the next sample to be studied.

In initial studies of this sample the usual chemical separation scheme was applied. Because of the reduced gross gamma activity in the sample, a volume of one liter was chosen.

Gamma spectral and decay data for the Silver Group separated from one of the initial samples collected from the anion exchanger effluent are shown in Figures 40 and 41. The two predominant radioactive nuclides present in this sample are 8.05 d I^{131} and 21 h I^{133}, the former being characterized by the 0.37 and 0.63 Mev gamma peaks and the latter by the 0.53 and 0.85 Mev gamma peaks. The initial rapid decay of the sample indicates also the presence

TABLE IV

Radioactive Nuclides Identified in the Effluent from the LITR Cation Exchanger

Date of sample collection, 12-17-56

Chemical Group	Radioactive Nuclide	Half Life	Estimated Concentration (μc/ml)*
Silver	I^{131}	8.05 d	4×10^{-5}
	+ Xe^{131m1}	4.8×10^{-10} s	
	I^{132}	2.3 h	
	I^{133}	21 h	4×10^{-4}
	+ Xe^{133}	5.27 d	2×10^{-3}
	+ Cs^{133m}	6.0×10^{-9} s	
	I^{135}	6.7 h	
	+ Xe^{135}	9.2 h	
	+ Cs^{135m}	2.8×10^{-10} s	
Acid Sulfide	Tc^{99m}	6.0 h	4×10^{-4}
	Sb^{122}	2.8 d	2×10^{-5}
Hydroxide	Tc^{99m}	6.0 h	1×10^{-4}
	F^{18}	1.87 h	5×10^{-5}
	Pa^{233}	27.4 d	5×10^{-6}
Basic Sulfide	Tc^{99m}	6.0 h	5×10^{-5}
Alkaline Earth	Tc^{99m}	6.0 h	5×10^{-6}
	Pa^{233}	27.4 d	4×10^{-6}
	F^{18}	1.87 h	5×10^{-6}
Soluble	---	---	---

* At time of sample collection.

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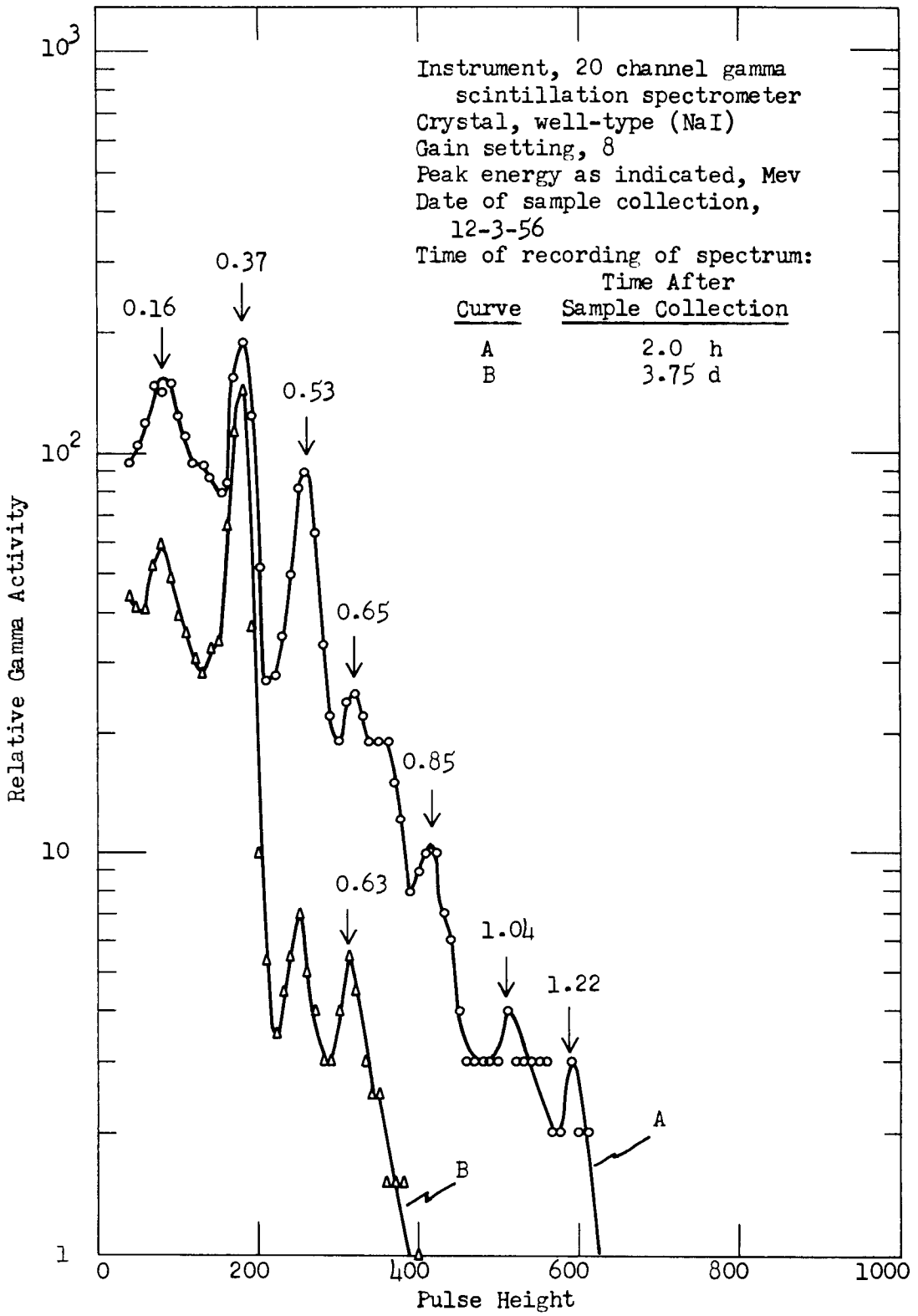

Fig. 40. Gamma Spectral Data for Effluent from LITR Anion Exchanger: Silver Group.

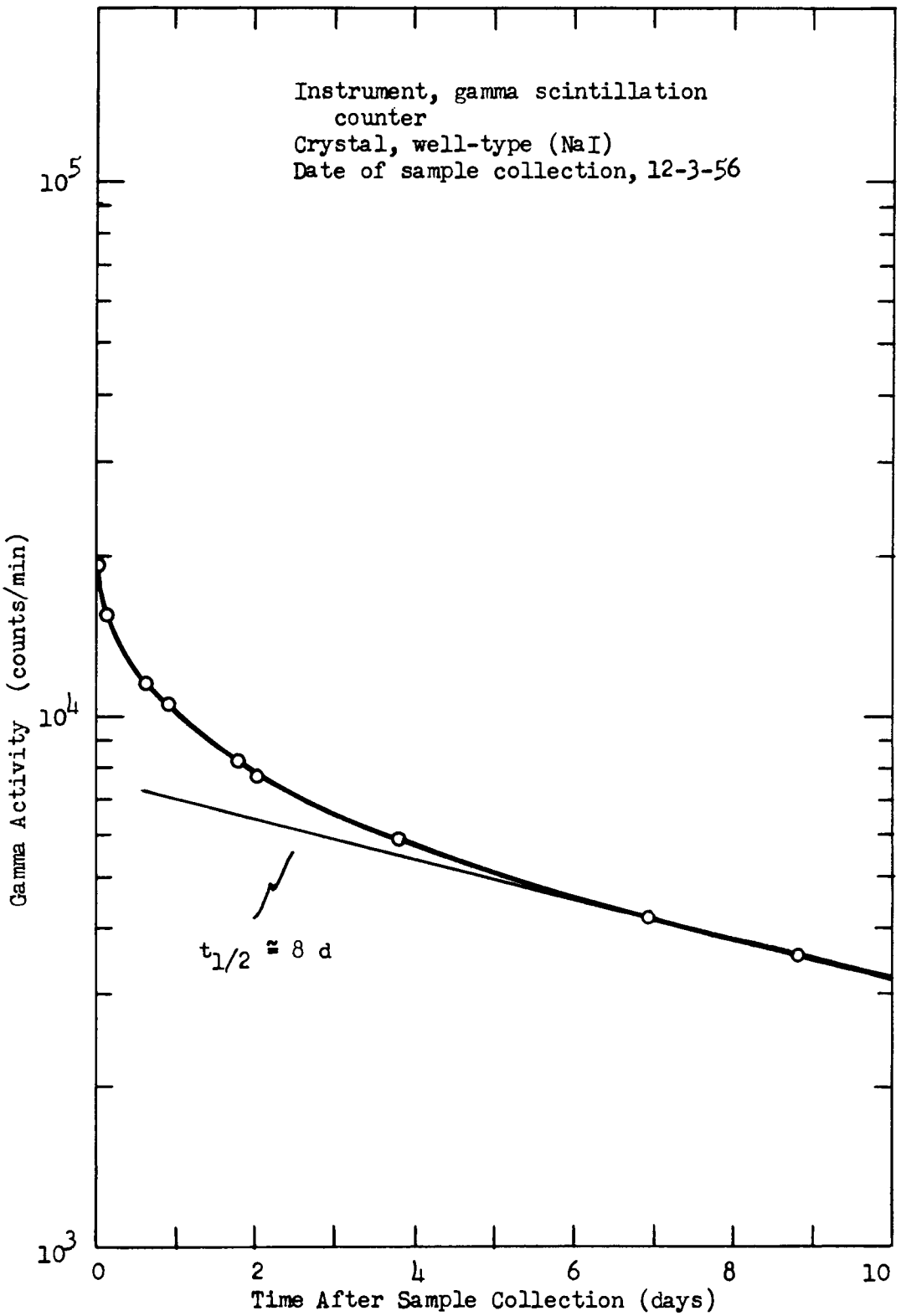

Fig. 41. Gross Gamma Decay for Effluent from LITR Anion Exchanger: Silver Group.

of 2.3 h I^{132}. This sample was unusual in that, even though the first spectrum was made only two hours after sample collection, I^{131} constituted a major portion of the total activity. In the same chemical group separated from the influent and effluent samples of the cation exchanger, I^{131} initially constituted only a minor fraction of the total activity.

The anion exchanger in use at this time was the smaller of the two previously described. The Silver Group separated from a second sample collected four weeks after placing the larger anion unit in operation revealed the gamma spectral data shown in Figure 42. Again, not all the iodine activity is being removed from the water by the column. However, in this second sample, 2.3 h I^{132} and 21 h I^{133} constitute the major activities, 8.05 d I^{131} becoming significant only after several days decay.

These data plus additional information obtained from a study of the anion resin (to be presented later in this report) strongly support the conclusion that iodine is loosely retained by the anion column. As a result, in an old bed the predominant radioactive material re-entering the cooling water stream at the bottom of the bed is I^{131}, the time of retention of the iodine on the bed being long in comparison with the half lives of the other iodine isotopes.

Such a phenomenon emphasizes the care that must be exercised in using isotopic ratios of a given radioactive element in, for example, the determination of the age of the fission products in a particular sample.

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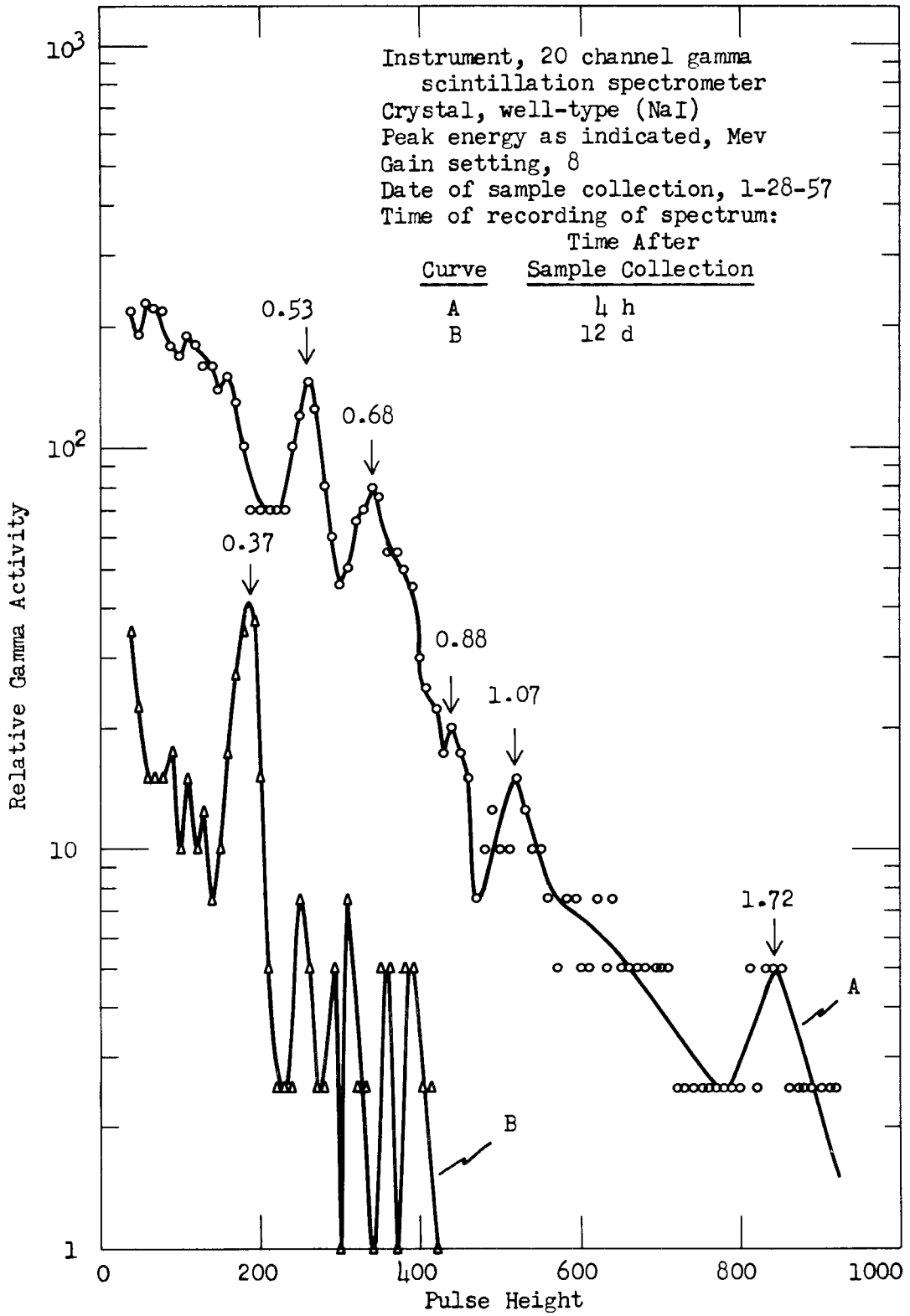

Fig. 42. Gamma Spectral Data for Effluent from LITR Anion Exchanger: Silver Group.

The chemical separation scheme, previously outlined, proved inadequate for positive identification of the radioactive nuclides remaining in the water sample. However, it did prove useful for making assumptions regarding the nuclides present. These assumptions were then either confirmed or denied through specific chemical separations for the particular nuclides in question.

Gross gamma measurements of the supernatant remaining after separation of the Silver Group indicated that the radioactive isotopes of iodine made up a large portion of the total radioactive material in the anion exchanger effluent. Therefore, for subsequent studies, samples of two liter volume were acidified with hydrochloric acid and evaporated to concentrate the remaining radioactive material.

Gamma spectral and decay data for the Acid Sulfide Group are shown in Figures 43 and 44. The 0.51, 1.04 and 1.36 Mev gamma peaks shown in the fresh sample represent 12.8 h Cu^{64}. The long-lived 0.57 Mev gamma peak is indicative of 2.8 d Sb^{122}. The 0.23 and 0.14 Mev gamma peaks are possibly indicative of 77 h Te^{132} and 6.0 h Tc^{99m}, respectively.

A tellurium separation of a similarly evaporated sample yielded the gamma spectral and decay data presented in Figures 45 and 46. These data confirm the presence of Te^{132}. The increase in gross gamma activity during the first day following the separation is due to growth of the 2.3 h I^{132} daughter activity. The separation of tellurium was accomplished through precipitation of elementary tellurium by reduction with sulfurous acid in 3 molar

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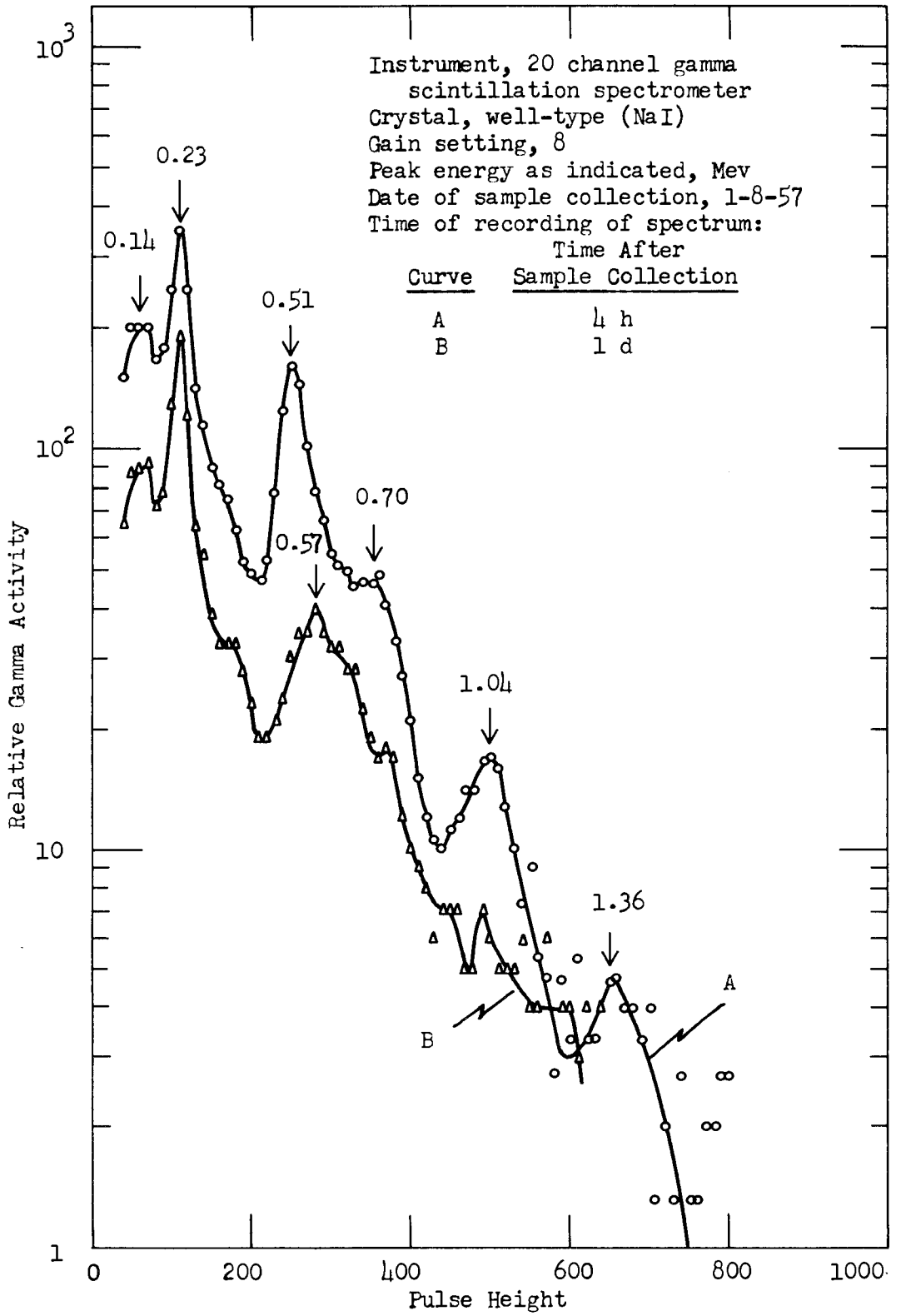

Fig. 43. Gamma Spectral Data for Effluent from LITR Anion Exchanger: Acid Sulfide Group.

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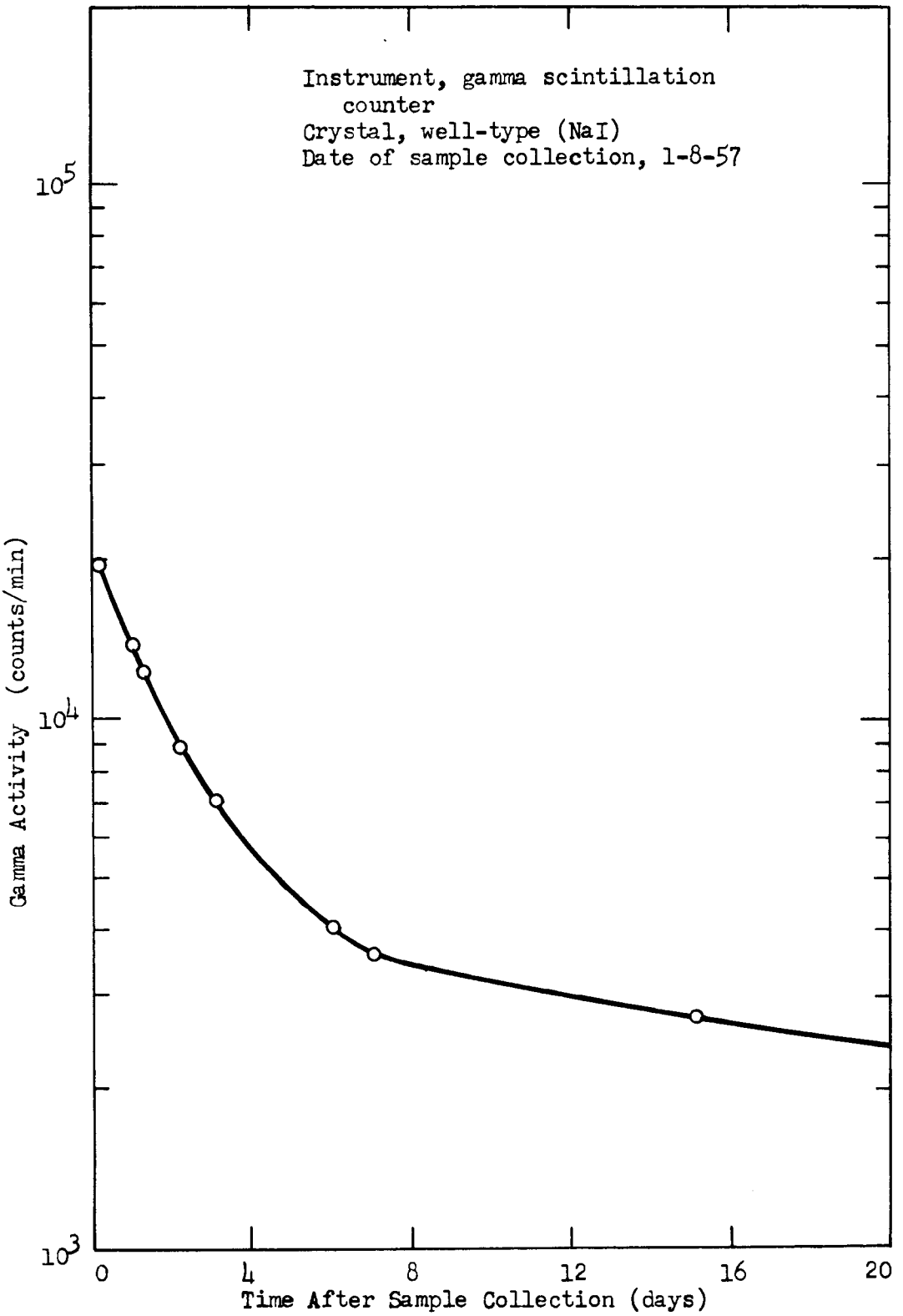

Fig. 44. Gross Gamma Decay for Effluent from LITR Anion Exchanger: Acid Sulfide Group.

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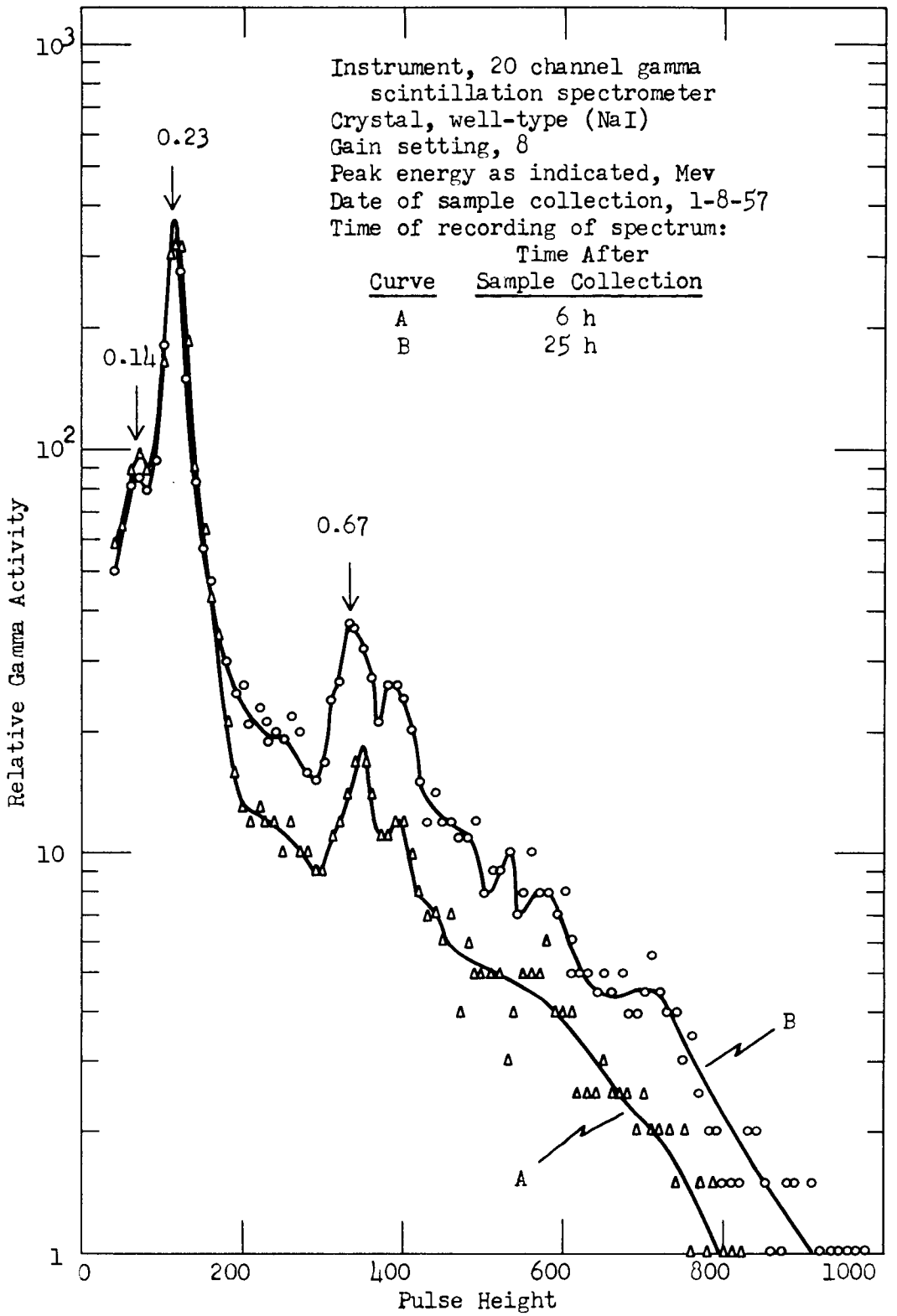

Fig. 45. Gamma Spectral Data for Effluent from LITR Anion Exchanger: Tellurium Separation.

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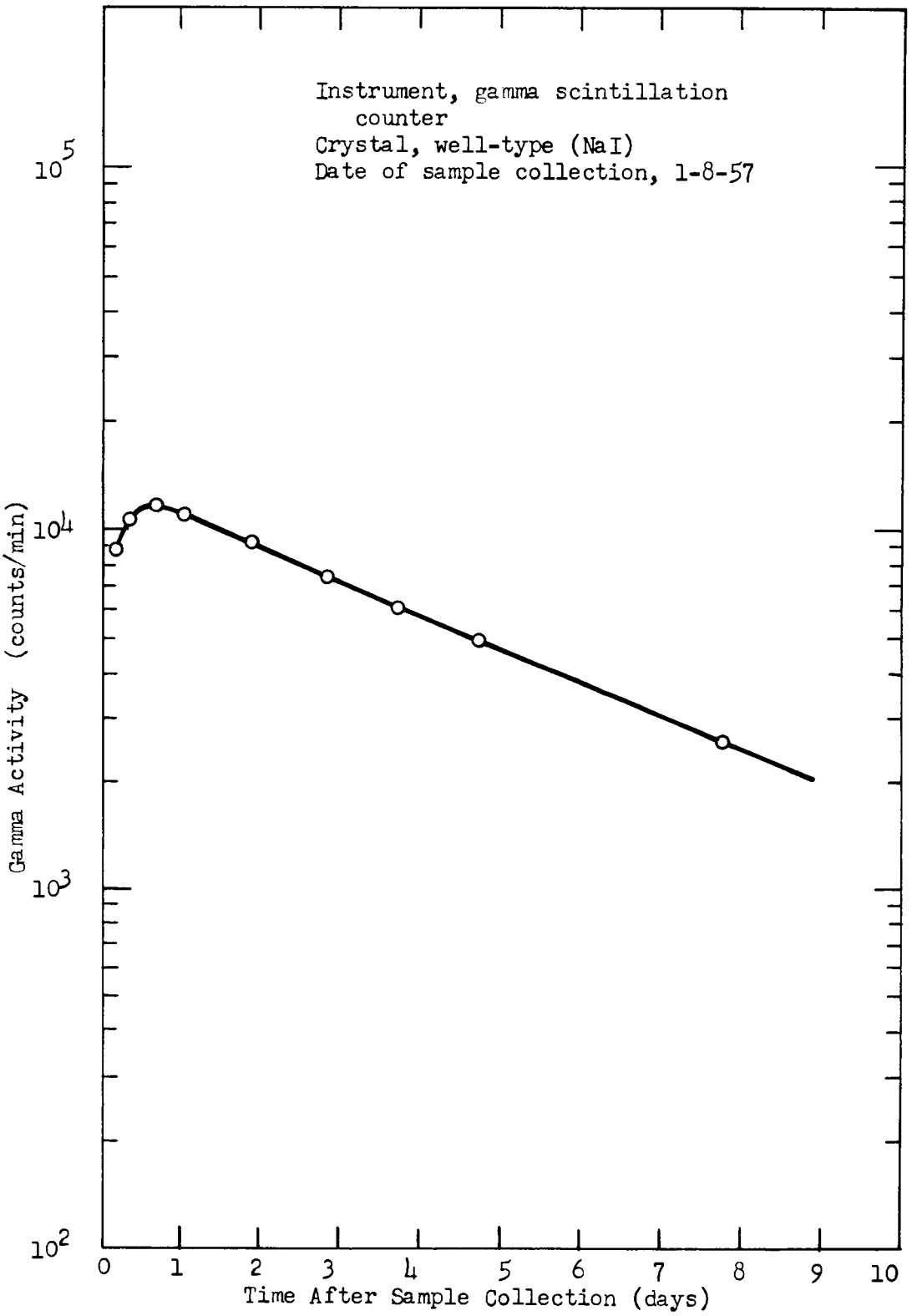

Fig. 46. Gross Gamma Decay for Effluent from LITR Anion Exchanger: Tellurium Separation.

hydrochloric acid. Volatilization of the sample with hydrobromic acid and scavenging with ferric hydroxide removed major contaminants (Hillebrand, et al., 1953).

The presence of 67 h Mo^{99} plus its daughter product 6.0 h Tc^{99m} was confirmed by a molybdenum separation on a similar sample. A portion of the spectral data showing the growth of the 0.14 Mev peak of the Tc^{99m} daughter is shown in Figure 47. Molybdenum was precipitated from a dilute acid solution of evaporated cooling water by alpha-benzoinoxime (Hillebrand, et al., 1953). Niobium would have been precipitated by this same procedure. However, gamma spectral studies failed to show its presence.

Gamma spectral and decay data for the Hydroxide Group showed the possible presence in the sample of 32 d Ce^{141} and 33 h Ce^{143}. Gamma spectral and decay data for a cerium separation on a similar sample are shown in Figures 48 and 49. These data confirm the presence of Ce^{141} and Ce^{143} in the anion exchanger effluent, Ce^{143} being represented by the 0.29, 0.64, 0.68 and 0.76 Mev gamma peaks and Ce^{141} being represented by the 0.14 Mev peak. Radioactive cerium was initially removed from the sample with the other rare earths by non-isotopic precipitation with lanthanum fluoride (Hillebrand, et al., 1953). The lanthanum fluoride was then dissolved, the cerium carrier added and the cerium carrier plus cerium radionuclides separated as the iodate.

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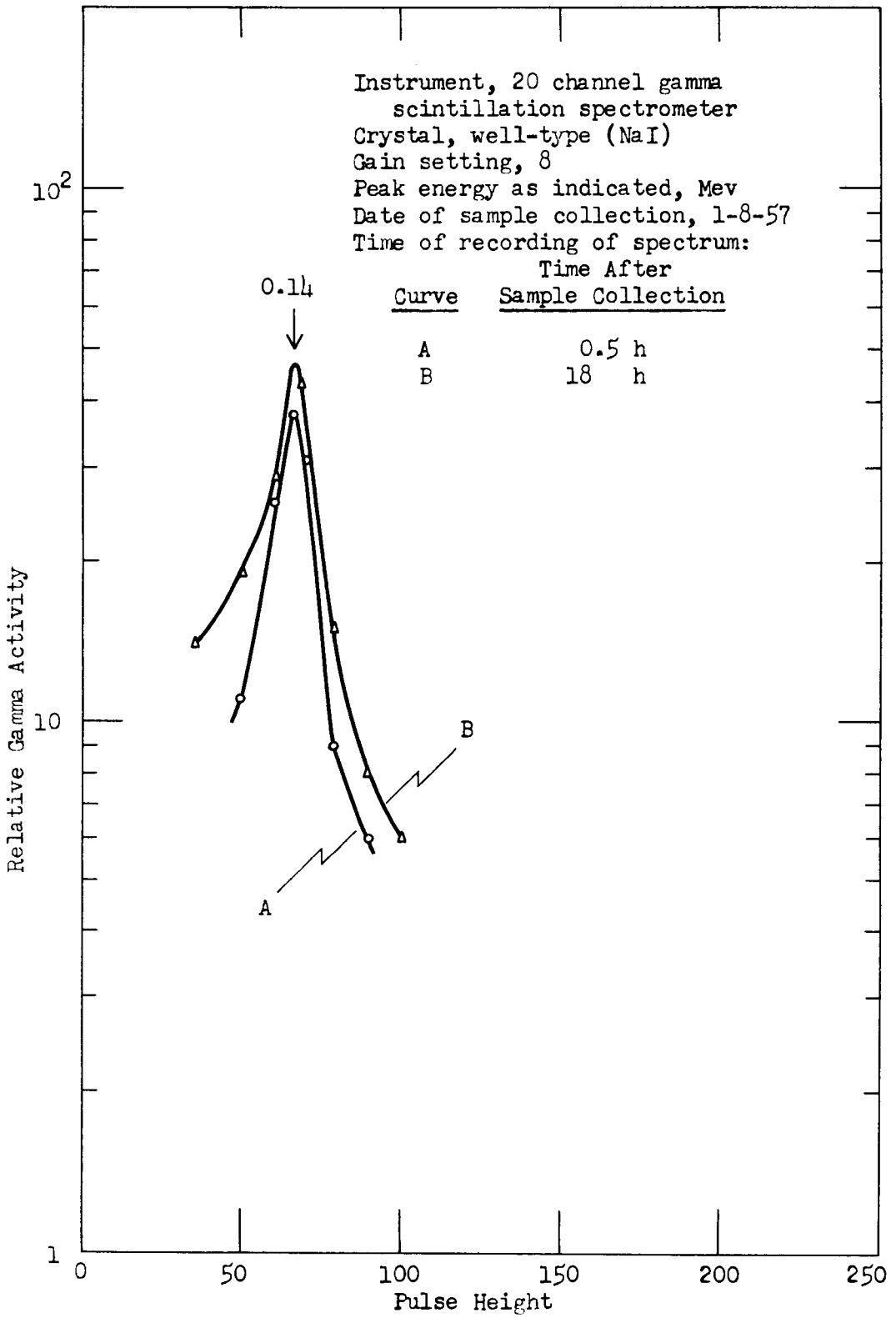

Fig. 47. Gamma Spectral Data for Effluent from LITR Anion Exchanger: Molybdenum Separation.

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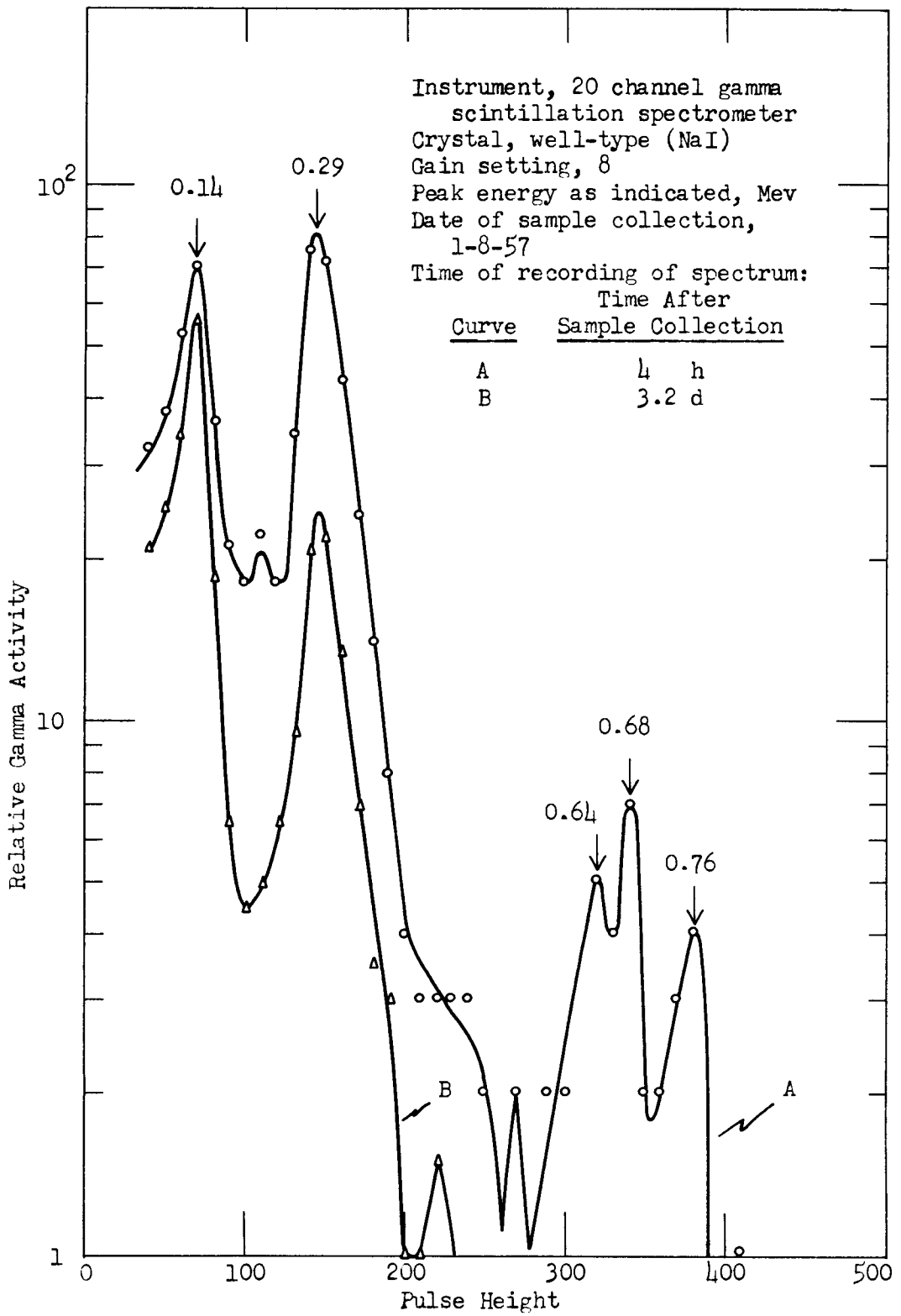

Fig. 48. Gamma Spectral Data for Effluent from LITR Anion Exchanger: Cerium Separation.

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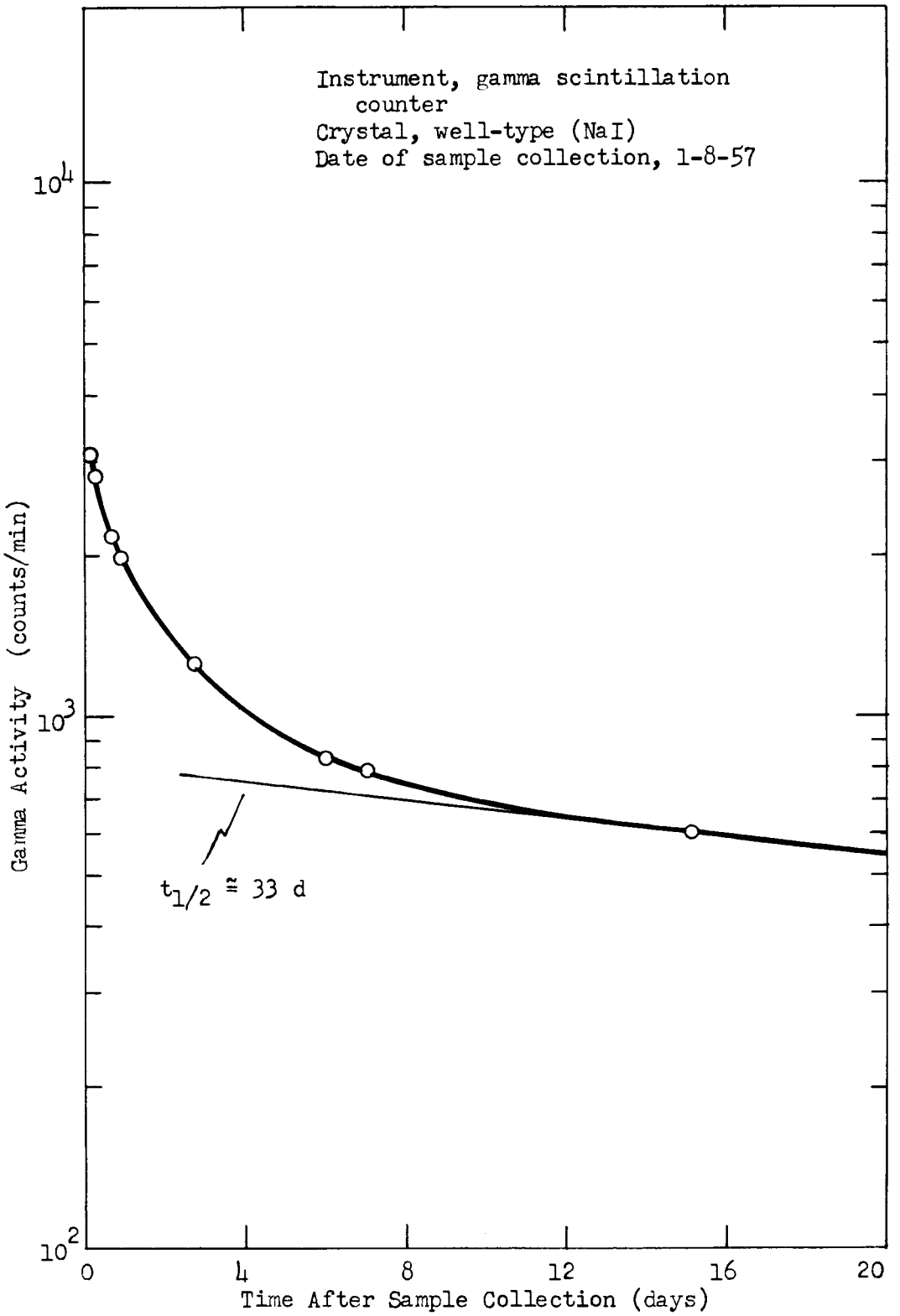

Fig. 49. Gross Gamma Decay for Effluent from LITR Anion Exchanger: Cerium Separation.

An iron Hydroxide Group precipitation of the remaining sample indicated the presence of 27 d Cr^{51}. Gamma spectral and decay data for this sample are shown in Figures 50 and 51. Chromium-51 is represented by the 0.32 Mev gamma peak. The 1.10 and 1.30 Mev peaks are indicative of 45 d Fe^{59}. The 0.64 and 0.82 Mev peaks possibly represent I^{132} and Mn^{56}, respectively. However, their presence was not confirmed.

The quantity of radioactive material in subsequent group separations following the usual chemical scheme was insignificant.

A summary of the radioactive nuclides identified in the anion exchanger effluent and their relative concentrations is given in Table V.

E. Mixed-Bed Exchanger Effluent

The next stage in the cooling water system was the mixed-bed exchanger, which receives water directly from the effluent side of the anion column. The effluent from the mixed-bed exchanger represented the final purified product being returned to the main cooling system. The radioactive nuclides in this sample would be those which the described purification system failed to remove effectively.

Because of an anticipated low concentration of radioactive material, samples of two liter volume were collected. A Silver Group precipitated from the sample as collected (evaporation would cause the iodine to be lost) failed to show the presence of

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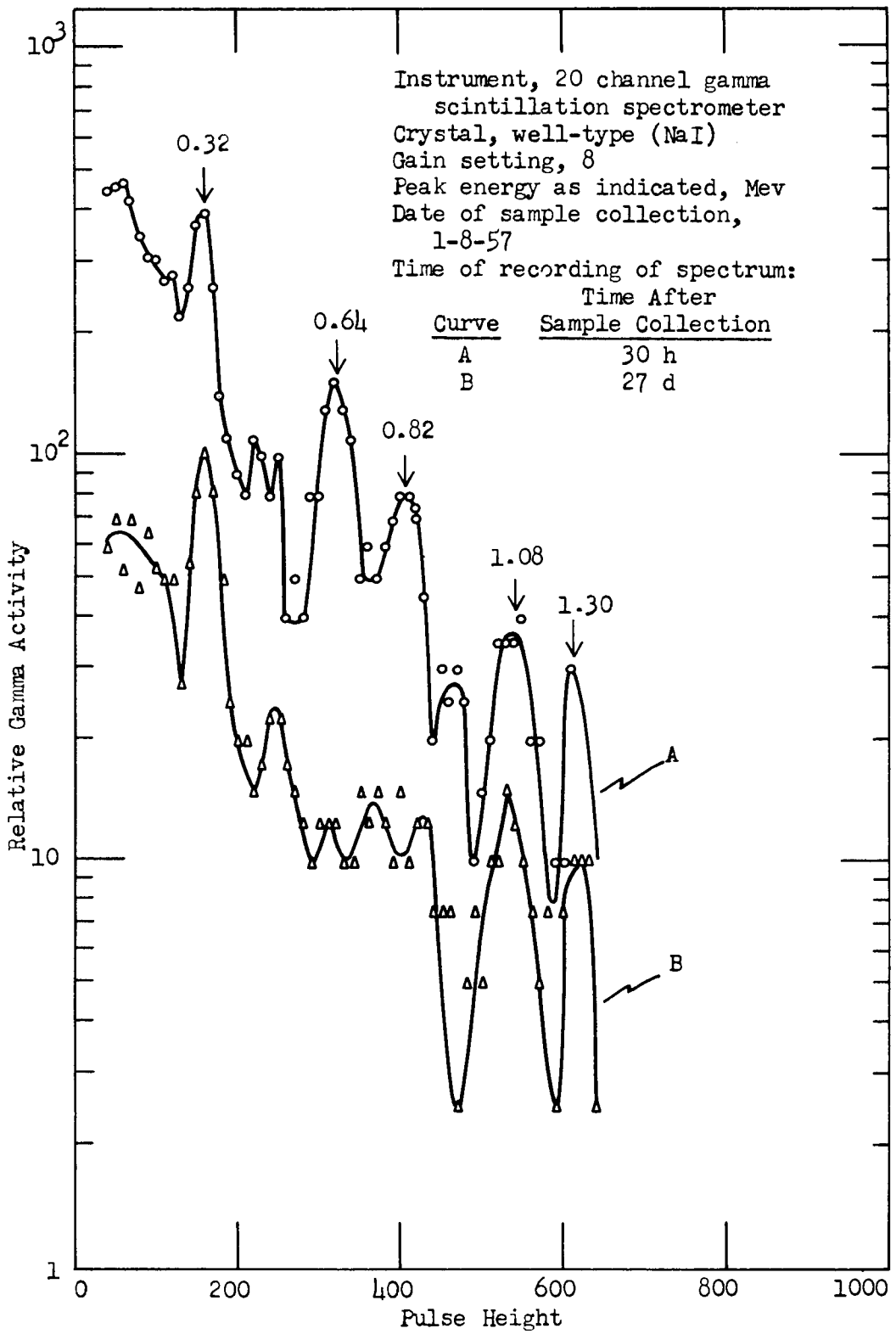

Fig. 50. Gamma Spectral Data for Effluent from LITR Anion Exchanger: Hydroxide Group.

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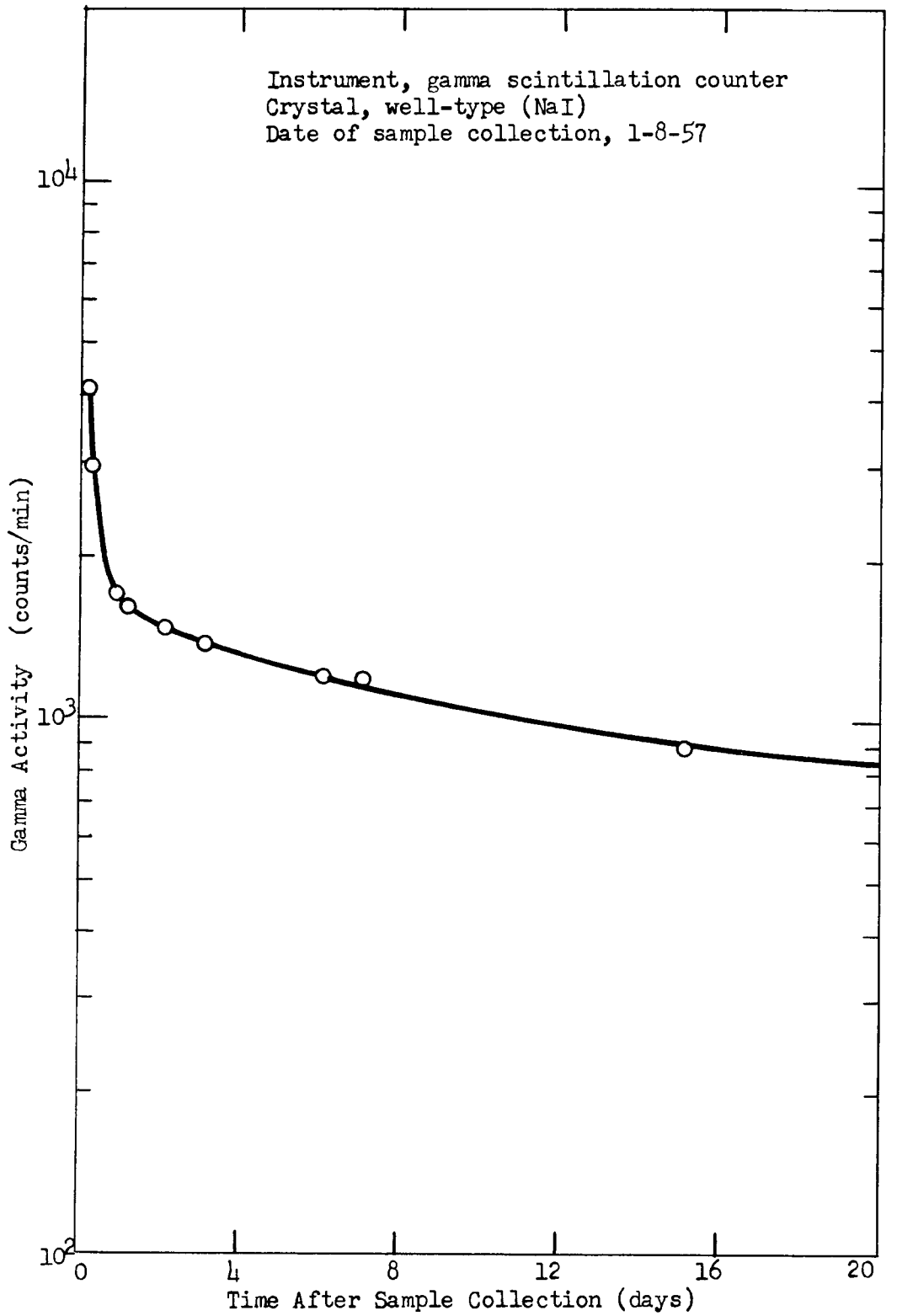

Fig. 51. Gross Gamma Decay for Effluent from LITR Anion Exchanger: Hydroxide Group.

TABLE V

Radioactive Nuclides Identified in the Effluent from the LITR Anion Exchanger

Date of sample collection, 1-8-57, except as noted otherwise

Chemical Group	Radioactive Nuclide	Half Life	Estimated Concentration (μc/ml)*
Silver	I^{131}	8.05 d	3×10^{-7}
	+ Xe^{131m1}	4.8×10^{-10} s	
	I^{132}	2.3 h	2×10^{-5}
	I^{133}	21 h	1×10^{-5}
			3×10^{-5}**
	I^{131}	8.05 d	2×10^{-5}
	I^{132}	2.3 h	5×10^{-6}
	I^{133}	21 h	2×10^{-5}
			4×10^{-5}***
Acid Sulfide	Cu^{64}	12.8 h	1×10^{-5}
	Te^{132}	77 h	1×10^{-5}
	+ I^{132}	2.3 h	
	Mo^{99}	67 h	3×10^{-6}
	+ Tc^{99m}	6.0 h	
	Sb^{122}	2.8 d	2×10^{-6}
Hydroxide	Ce^{141}	32 d	1×10^{-6}
	Ce^{143}	33 h	3×10^{-6}
	+ Pr^{143}	13.8 d	
	Cr^{51}	27 d	1×10^{-6}
	Fe^{59}	45 d	5×10^{-7}
Basic Sulfide	---	---	---
Alkaline Earth	---	---	---
Soluble	---	---	---

* At the time of sample collection.

** Sample collected on 1-28-57; larger anion exchanger in use.

*** Sample collected on 12-3-56; smaller anion exchanger in use.

significant iodine activity. For further studies, samples of two liter volume were acidified with hydrochloric acid and evaporated to approximately five milliliters. Subsequent group separations were then made on the evaporated sample.

Gamma spectral and decay data for the Acid Sulfide Group are shown in Figures 52 and 53. The predominant radionuclide in this group is the positron emitter 12.8 h Cu^{64}, as shown by the 0.51, 1.02 and 1.34 Mev gamma peaks. As in the spectral data for the same group from the anion exchanger effluent, 6.0 h Tc^{99m}, 77 h Te^{132} and 2.3 h I^{132} are in evidence. As before, specific chemical separations confirmed the presence of these nuclides.

The data for the Hydroxide Group showed evidence for 32 d Ce^{141} and 33 h Ce^{143}. A specific chemical separation confirmed these two nuclides. Gamma spectral and decay data for the separation are shown in Figures 54 and 55. The reason for the apparent difference in relative amounts of the two cerium isotopes in this sample and that from the anion exchanger effluent is due to the delay in obtaining the spectrum in the former.

The data for the Hydroxide Group after removal of the cerium again indicated the presence of 27 d Cr^{51}. The radionuclide 2.3 h I^{132} was also indicated but not confirmed in this particular group.

The quantity of radioactive material in subsequent chemical groups separated by the usual chemical scheme was insignificant.

Table VI contains a summary of the radioactive nuclides identified in the effluent from the mixed-bed exchanger and their estimated concentrations at the time of sampling.

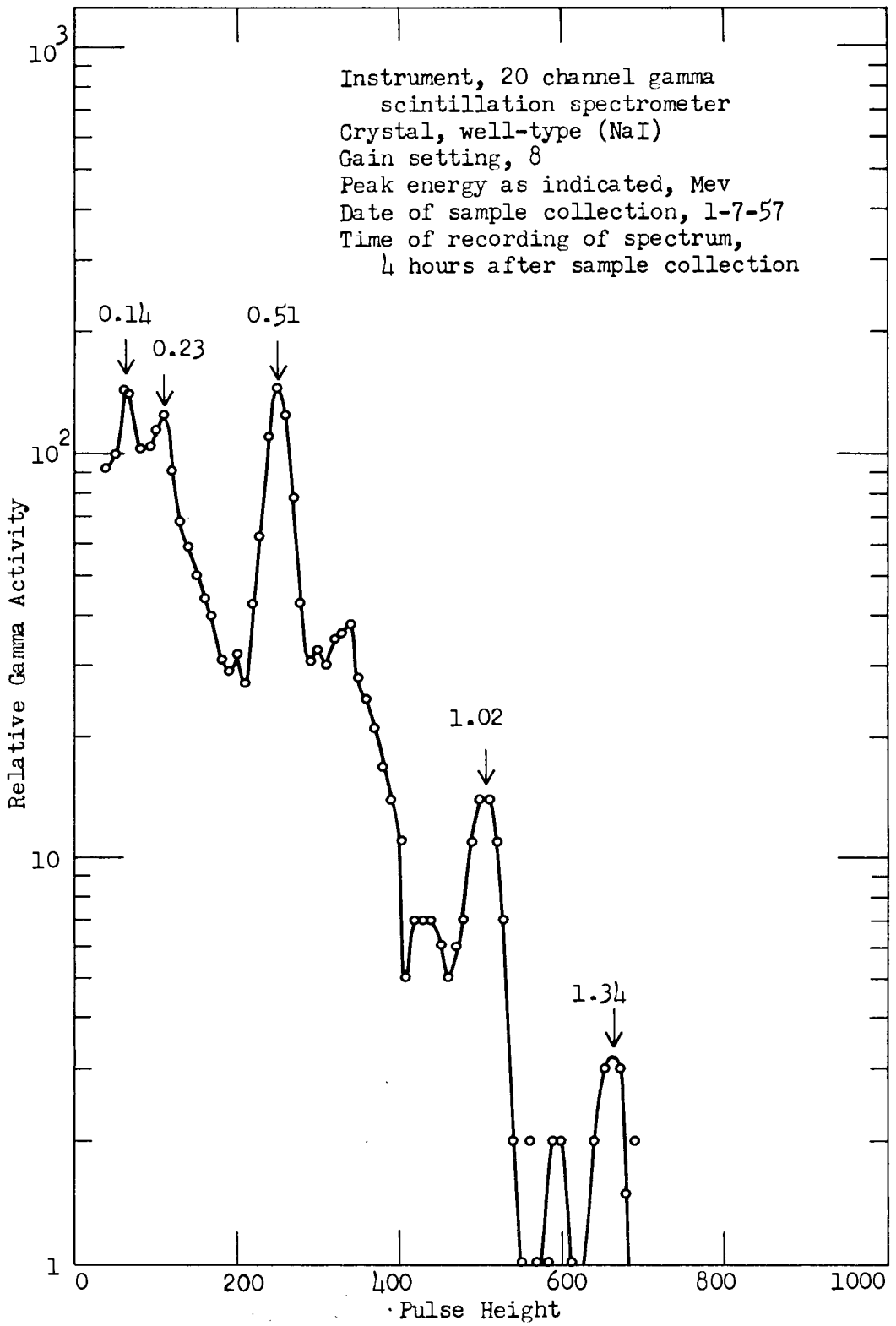

Fig. 52. Gamma Spectral Data for Effluent from LITR Mixed-Bed Exchanger: Acid Sulfide Group.

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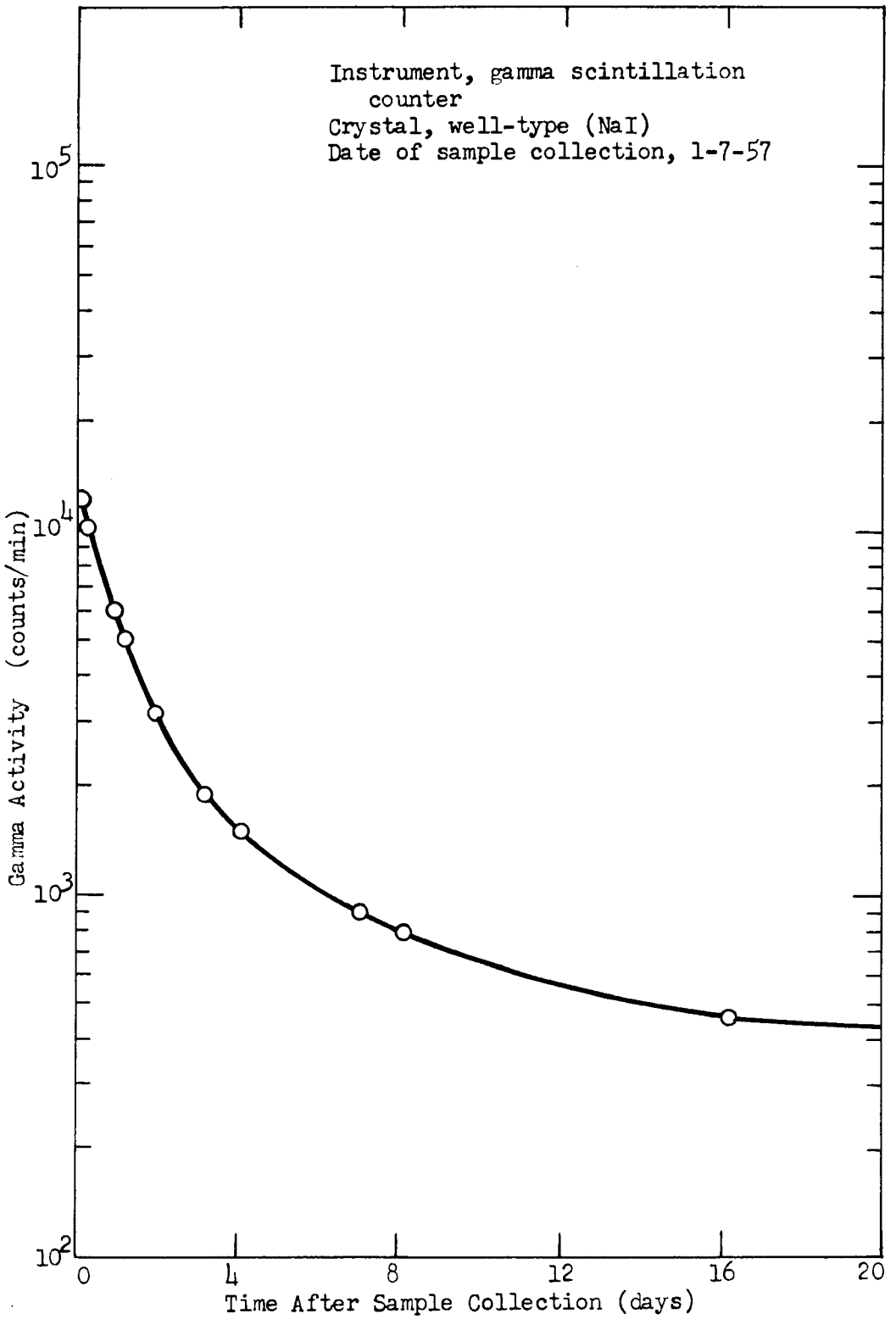

Fig. 53. Gross Gamma Decay for Effluent from LITR Mixed-Bed Exchanger: Acid Sulfide Group.

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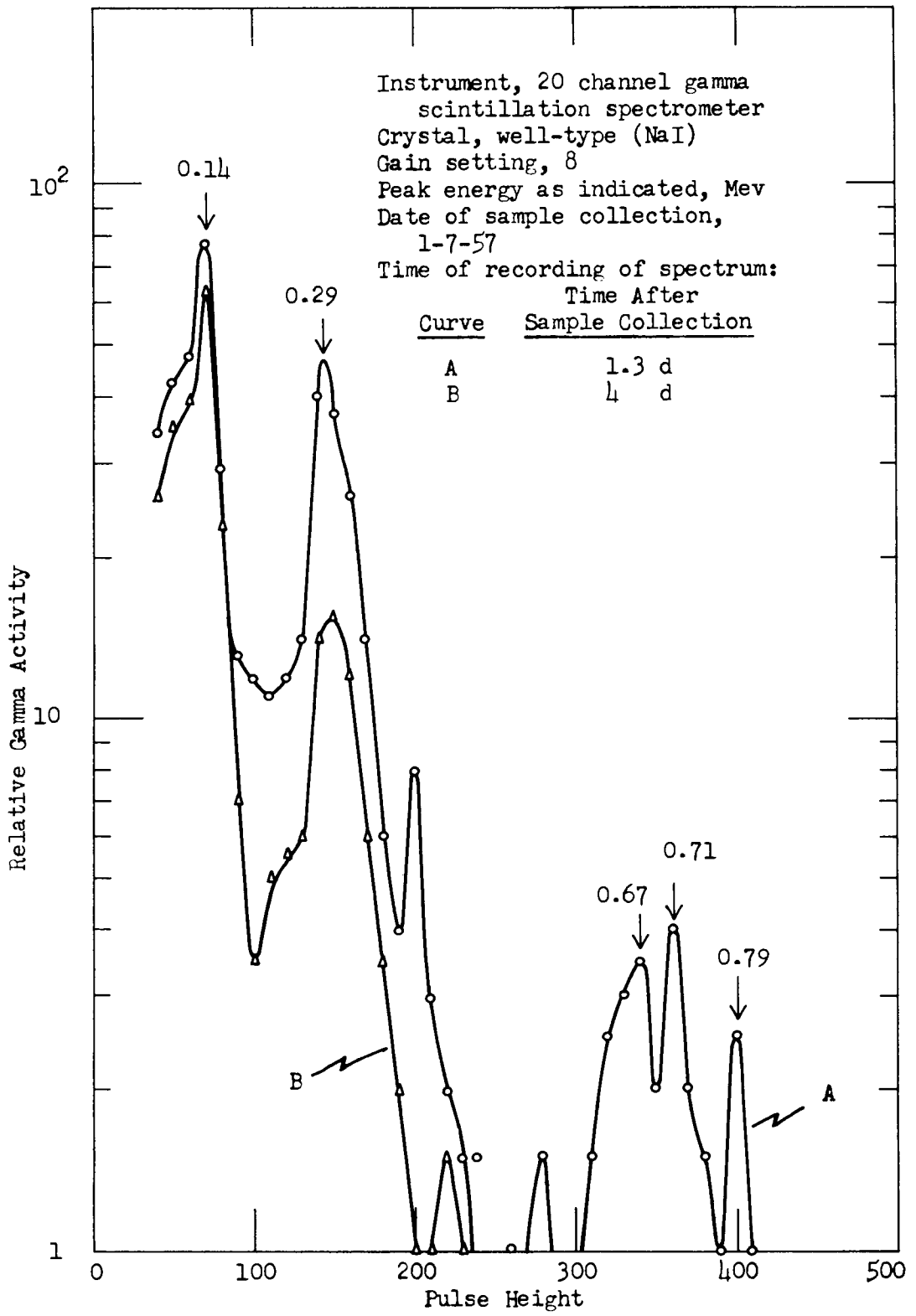

Fig. 54. Gamma Spectral Data for Effluent from LITR Mixed-Bed Exchanger: Cerium Separation.

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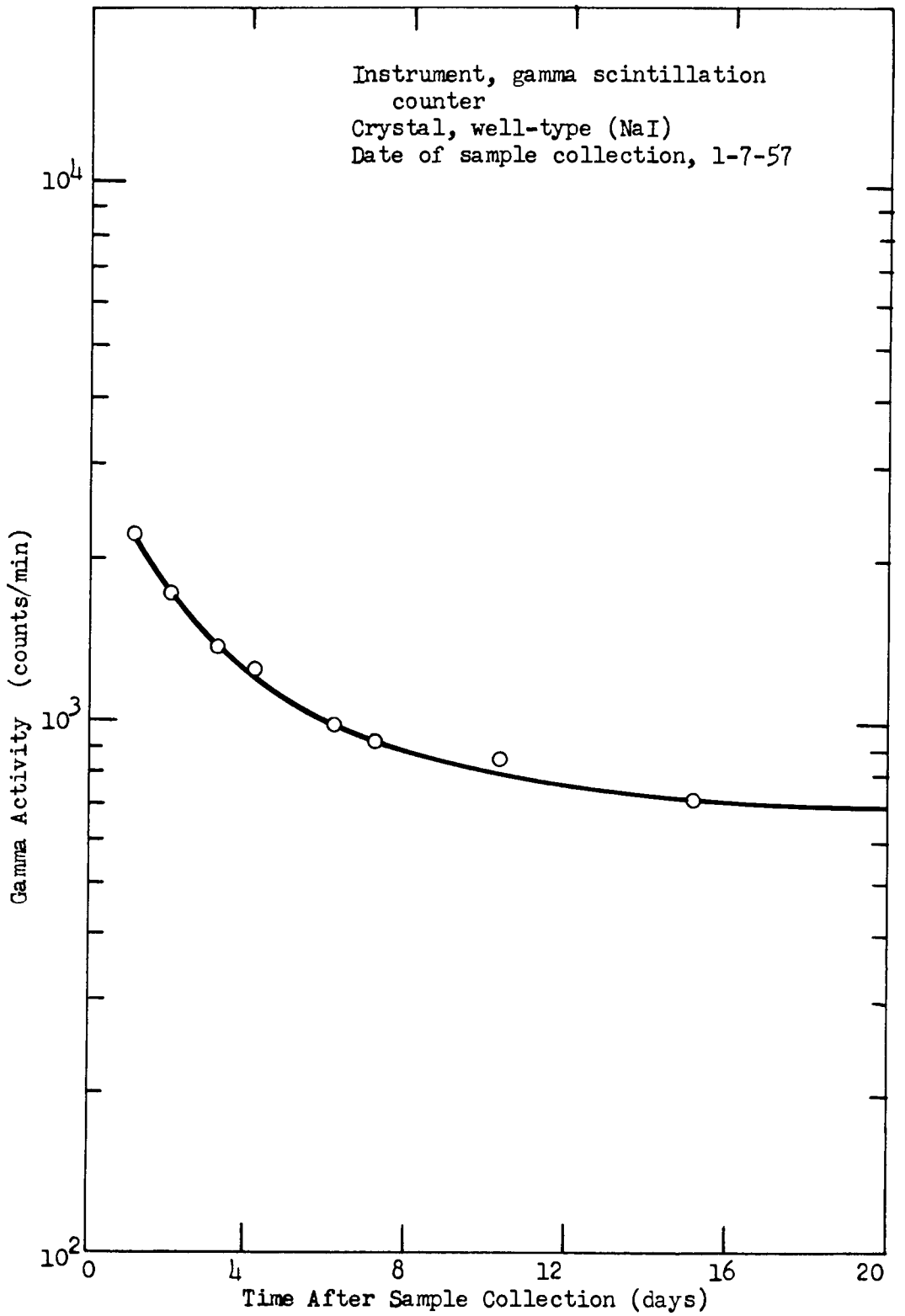

Fig. 55. Gross Gamma Decay for Effluent from LITR Mixed-Bed Exchanger: Cerium Separation.

TABLE VI

Radioactive Nuclides Identified in the Effluent from the LITR Mixed-Bed Exchanger

Date of sample collection, 1-7-57

Chemical Group	Radioactive Nuclide	Half Life	Estimated Concentration (μc/ml)*
Silver	I^{131}	8.05 d	$<1 \times 10^{-7}$
	I^{132}	2.3 h	
	I^{133}	21 h	
	I^{135}	6.7 h	
Acid Sulfide	Cu^{64}	12.8 h	1×10^{-5}
	Te^{132} + I^{132}	77 h 2.3 h	2×10^{-6}
	Mo^{99} + Tc^{99m}	67 h 6.0 h	3×10^{-6}
Hydroxide	Ce^{141}	32 d	1×10^{-6}
	Ce^{143} + Pr^{143}	33 h 13.8 d	4×10^{-6}
	Cr^{51}	27 d	1×10^{-6}
Basic Sulfide	---	---	---
Alkaline Earth	---	---	---
Soluble	---	---	---

* At time of sample collection.

F. Mixed-Bed Exchanger Effluent (Analysis for Stable Elements)

As an additional check on the purity of the cooling water being returned to the main system, a sample of the mixed-bed exchanger effluent was set aside to decay for three weeks and then analyzed for stable element concentration by neutron activation analysis.

A 55 milliliter aliquot of the decayed sample was placed in a polyethylene bottle and irradiated for 16 hours in the Hole 10 facility of the ORNL Graphite Reactor. The thermal flux was approximately 6.5×10^{11} neutrons·cm^{-2}·$second^{-1}$. The procedure followed was similar to that used in the analysis of the demineralized make-up water except that the 20 channel rather than the single channel gamma scintillation spectrometer was used to obtain the majority of the spectral data.

Gamma spectral and decay data for the Silver Group are shown in Figures 56 and 57. The 0.54, 0.77, 1.00 and 1.29 Mev peaks are characteristic of 35.9 h Br^{82}. The identification of Br^{82} is further substantiated by the gross gamma decay data. The decay data also show the probable presence of 4.6 h Br^{80m}. The 0.82 Mev peak represents 2.58 h Mn^{56}. Manganese is not normally found in the Silver Group. Perhaps the reason for its being found here is due to the fact that it was present in the sample as manganese dioxide and was carried by the silver precipitate (Wahl and Bonner, 1951).

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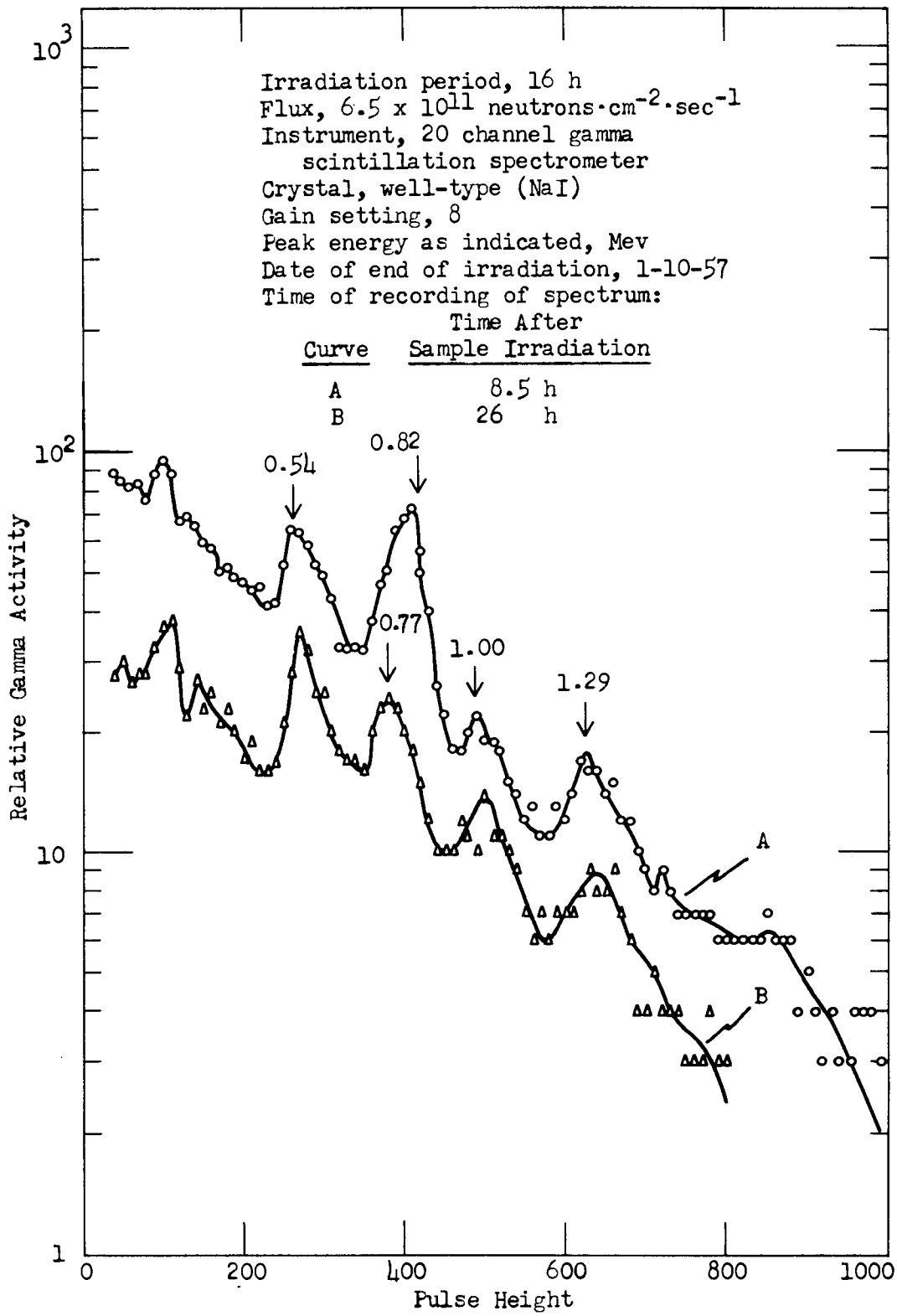

Fig. 56. Gamma Spectral Data for Neutron Irradiated Effluent from LITR Mixed-Bed Exchanger: Silver Group.

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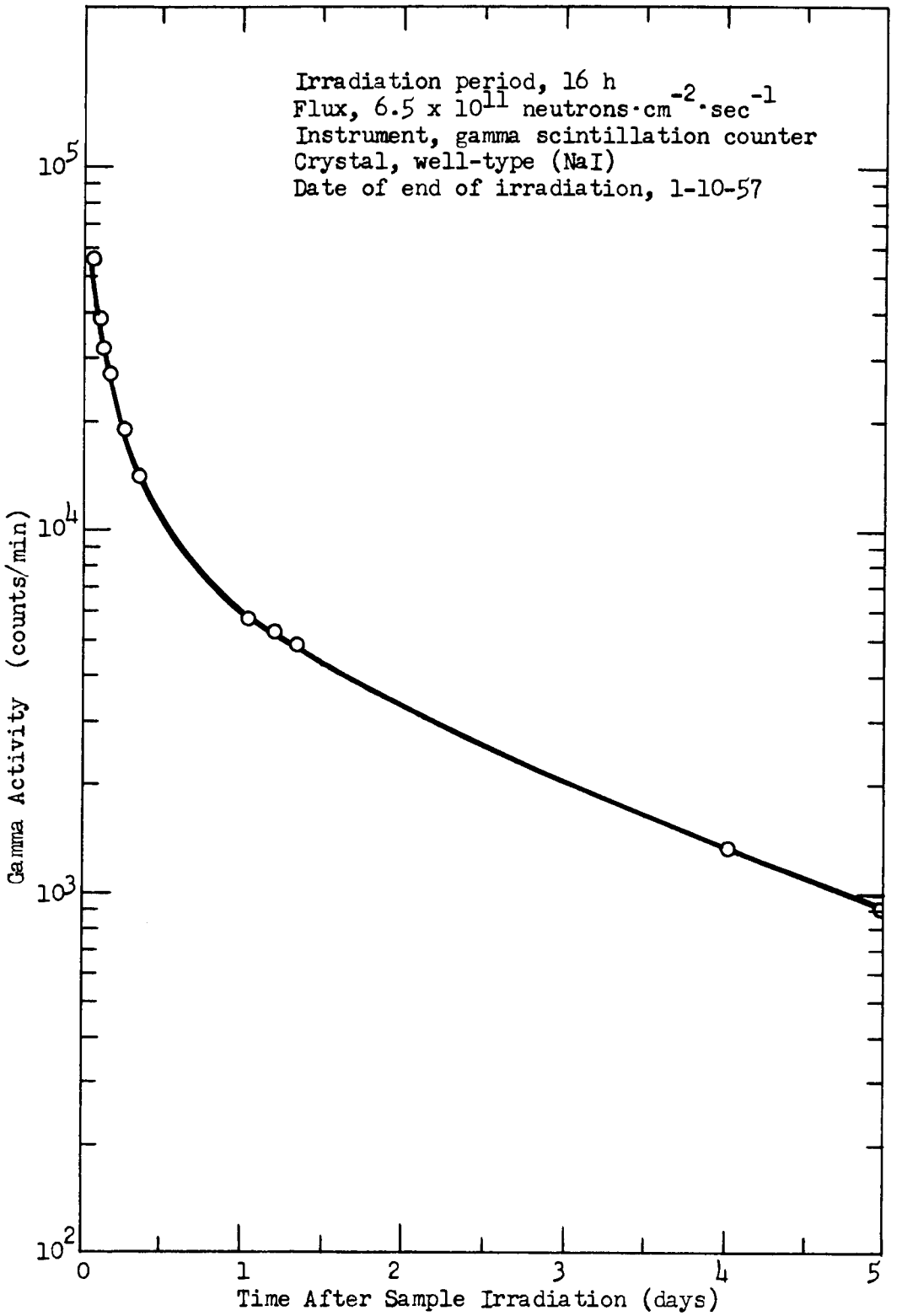

Fig. 57. Gross Gamma Decay for Neutron Irradiated Effluent from LITR Mixed-Bed Exchanger: Silver Group.

Data for the Acid Sulfide Group are shown in Figures 58 and 59. The predominant radioactive nuclide in this group was 12.8 h Cu^{64} as shown by the gross gamma decay data and the 0.51 and 1.01 Mev gamma peaks in the spectral data.

Gamma spectral and gross gamma decay data for the Hydroxide Group and the Alkaline Earth Group were very similar in that they both indicated the presence of 1.87 h F^{18}. Data for the Hydroxide Group are presented in Figures 60 and 61. The 0.51 Mev peak is typical of a positron emitter. The 1.05 Mev gamma peak represents "pile up". Data for the Alkaline Earth Group are not presented.

Gamma spectral and gross gamma decay data for the Basic Sulfide Group showed the presence of 2.58 h Mn^{56} in the activated sample. These data are shown in Figures 62 and 63.

Gamma spectral and gross gamma decay data for the Soluble Group showed the presence of 15.0 h Na^{24} in the activated sample. These data are shown in Figures 64 and 65. The spectral data for this group were obtained on the single channel gamma scintillation spectrometer.

Table VII contains a summary of the radioactive nuclides identified in the neutron irradiated effluent from the mixed-bed exchanger. Also listed are the estimated concentrations of the stable source elements of the given radionuclides.

It is interesting to note the similarity in radioactive nuclide composition of the activated make-up water and the activated mixed-bed exchanger effluent. The similarity is not surprising

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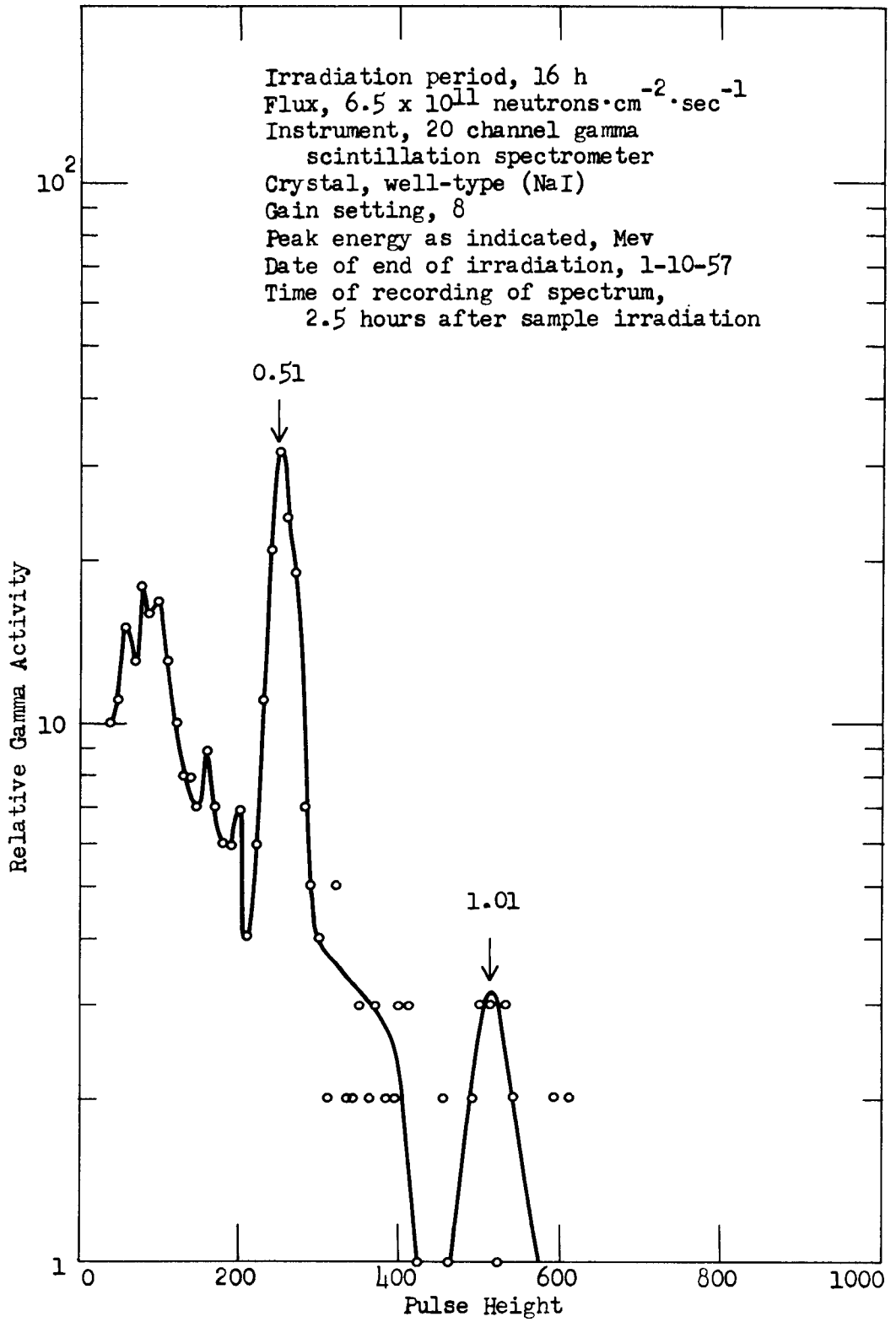

Fig. 58. Gamma Spectral Data for Neutron Irradiated Effluent from LITR Mixed-Bed Exchanger: Acid Sulfide Group.

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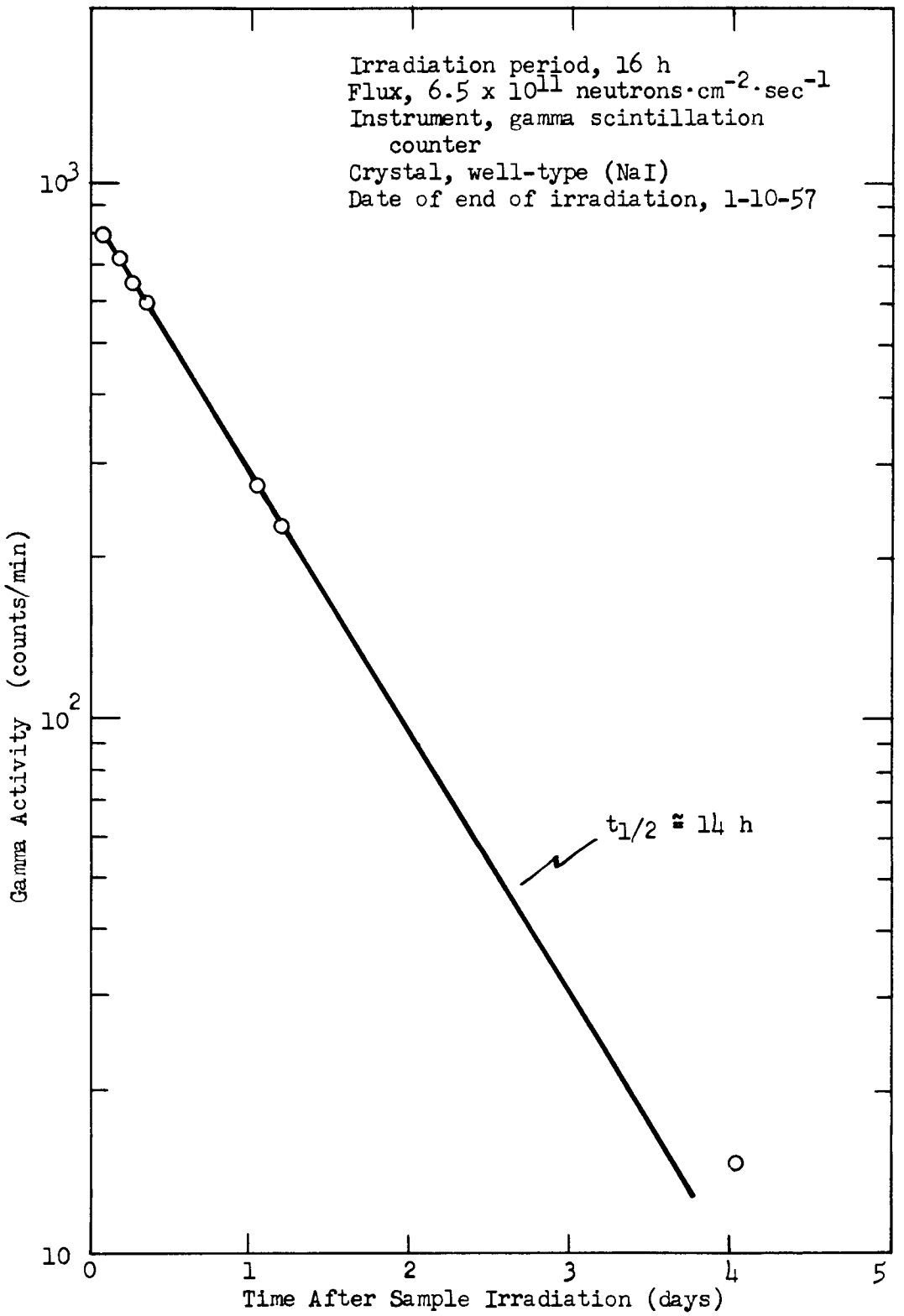

Fig. 59. Gross Gamma Decay for Neutron Irradiated Effluent from LITR Mixed-Bed Exchanger: Acid Sulfide Group.

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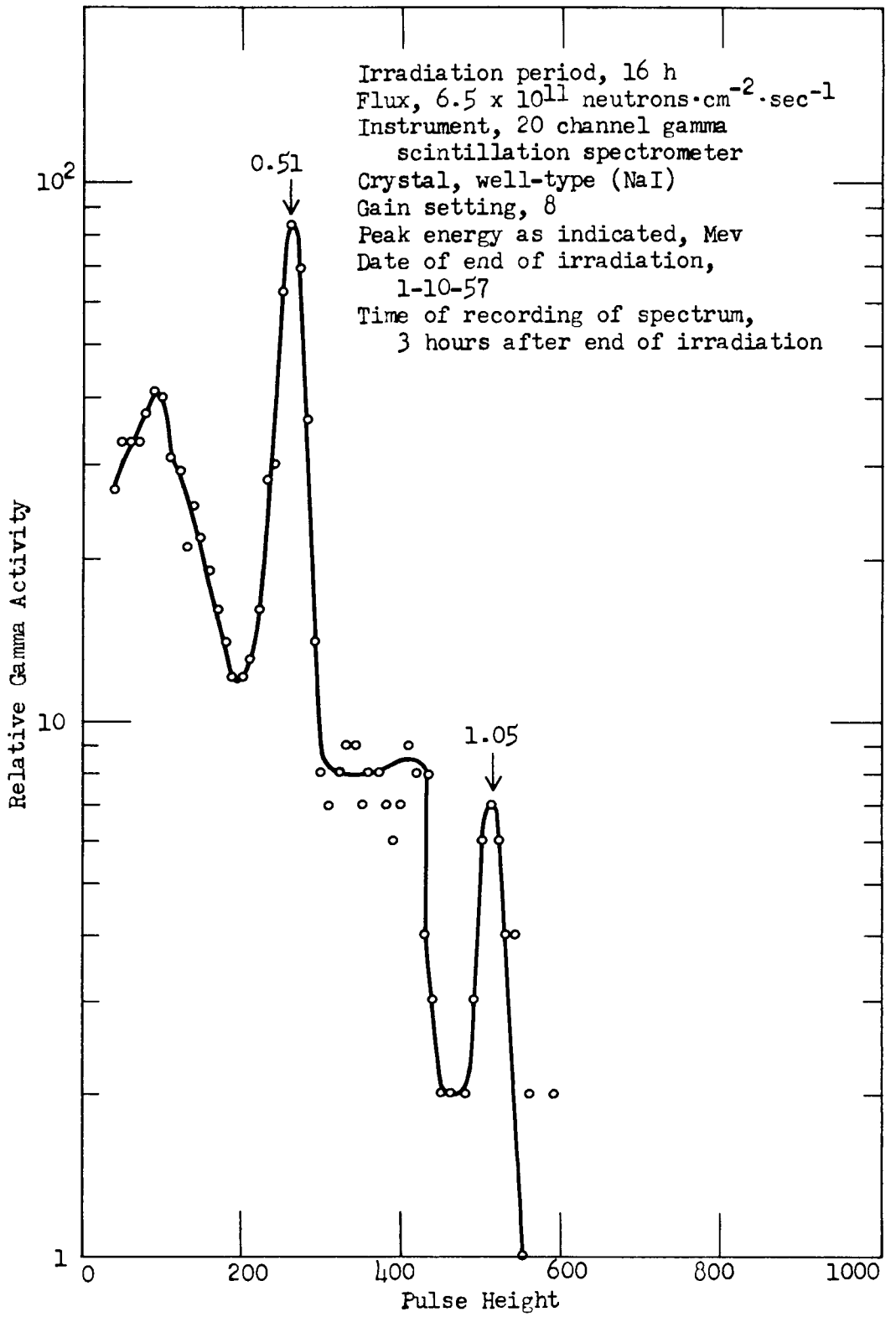

Fig. 60. Gamma Spectral Data for Neutron Irradiated Effluent from LITR Mixed-Bed Exchanger: Hydroxide Group.

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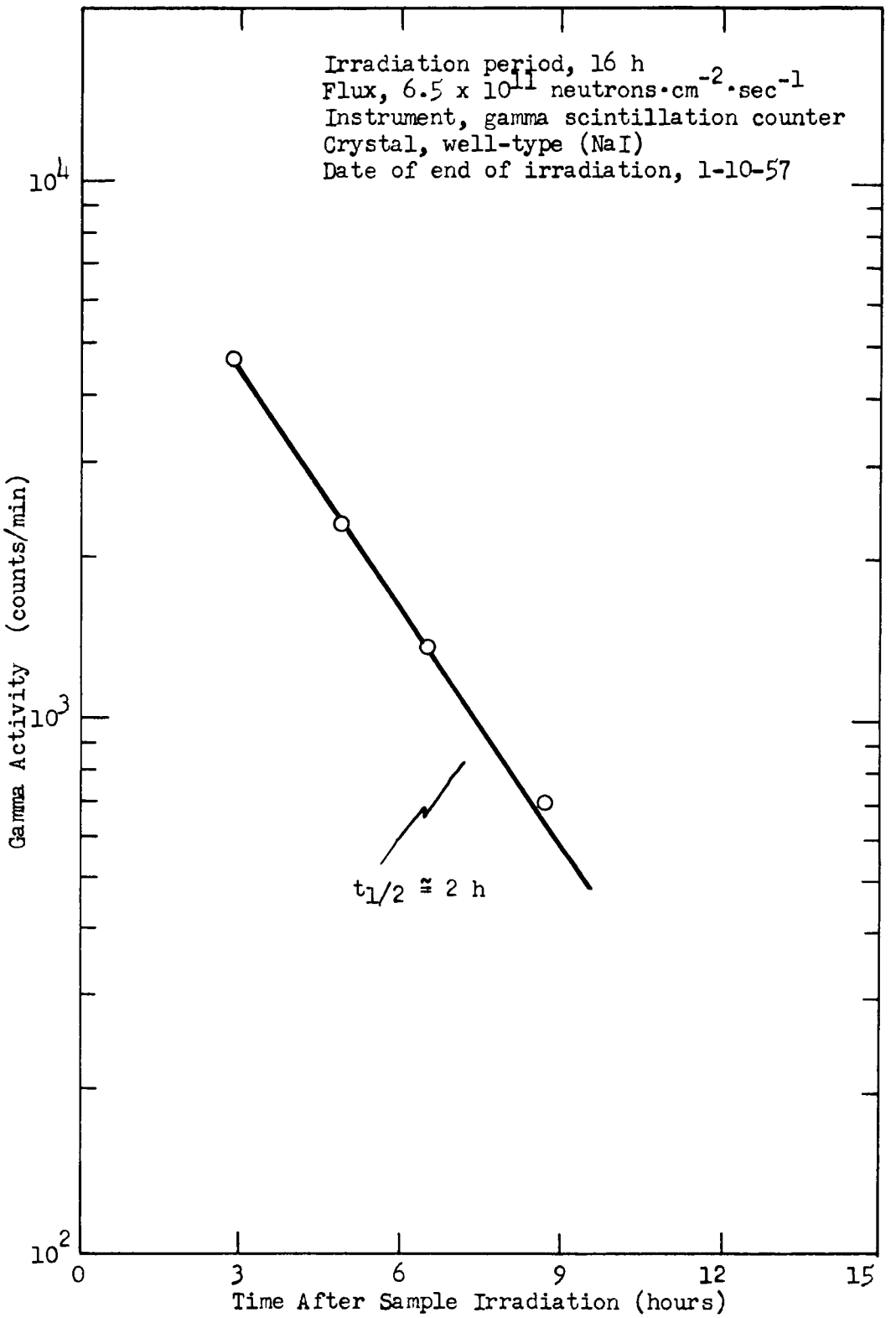

Fig. 61. Gross Gamma Decay for Neutron Irradiated Effluent from LITR Mixed-Bed Exchanger: Hydroxide Group.

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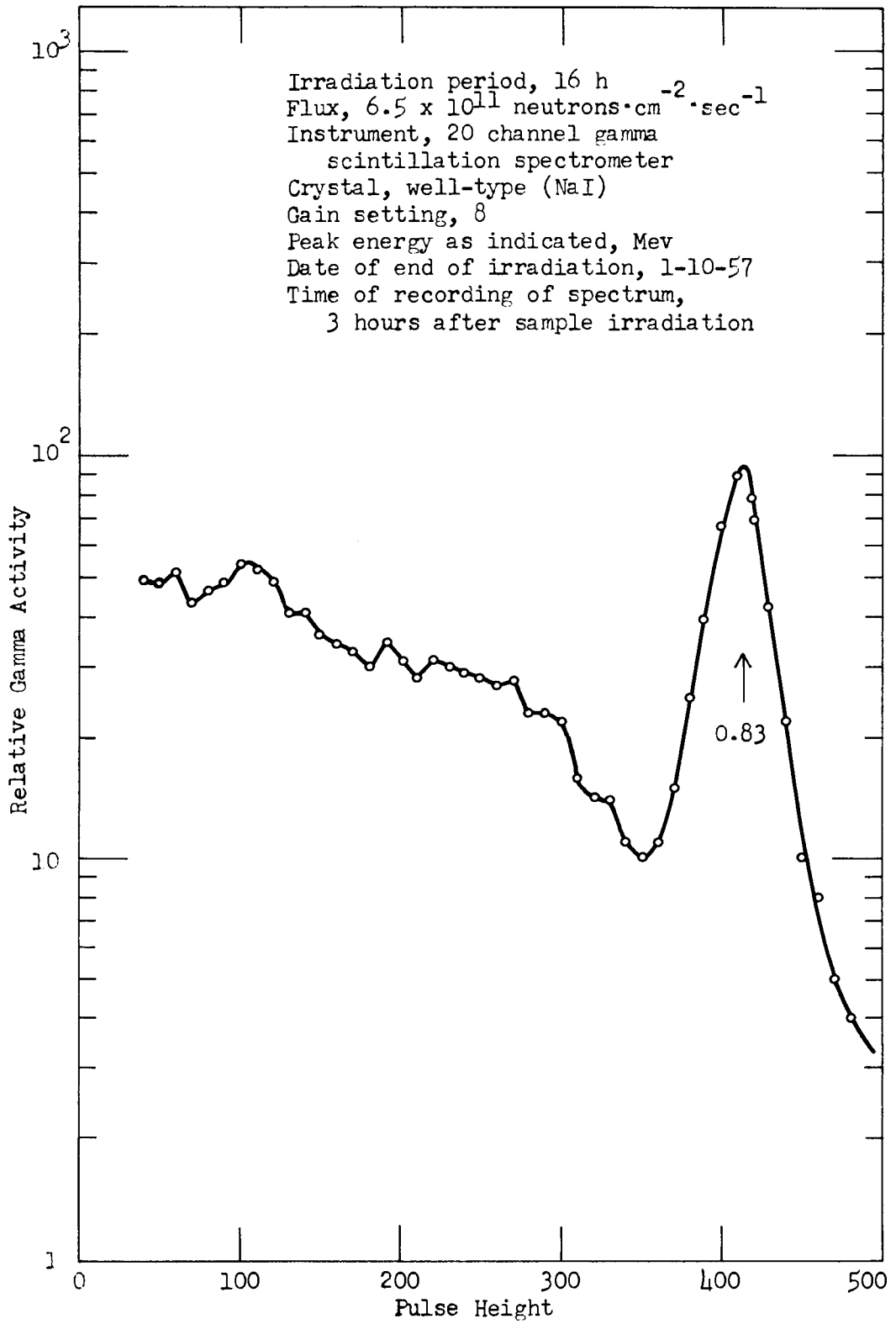

Fig. 62. Gamma Spectral Data for Neutron Irradiated Effluent from LITR Mixed Bed Exchanger: Basic Sulfide Group.

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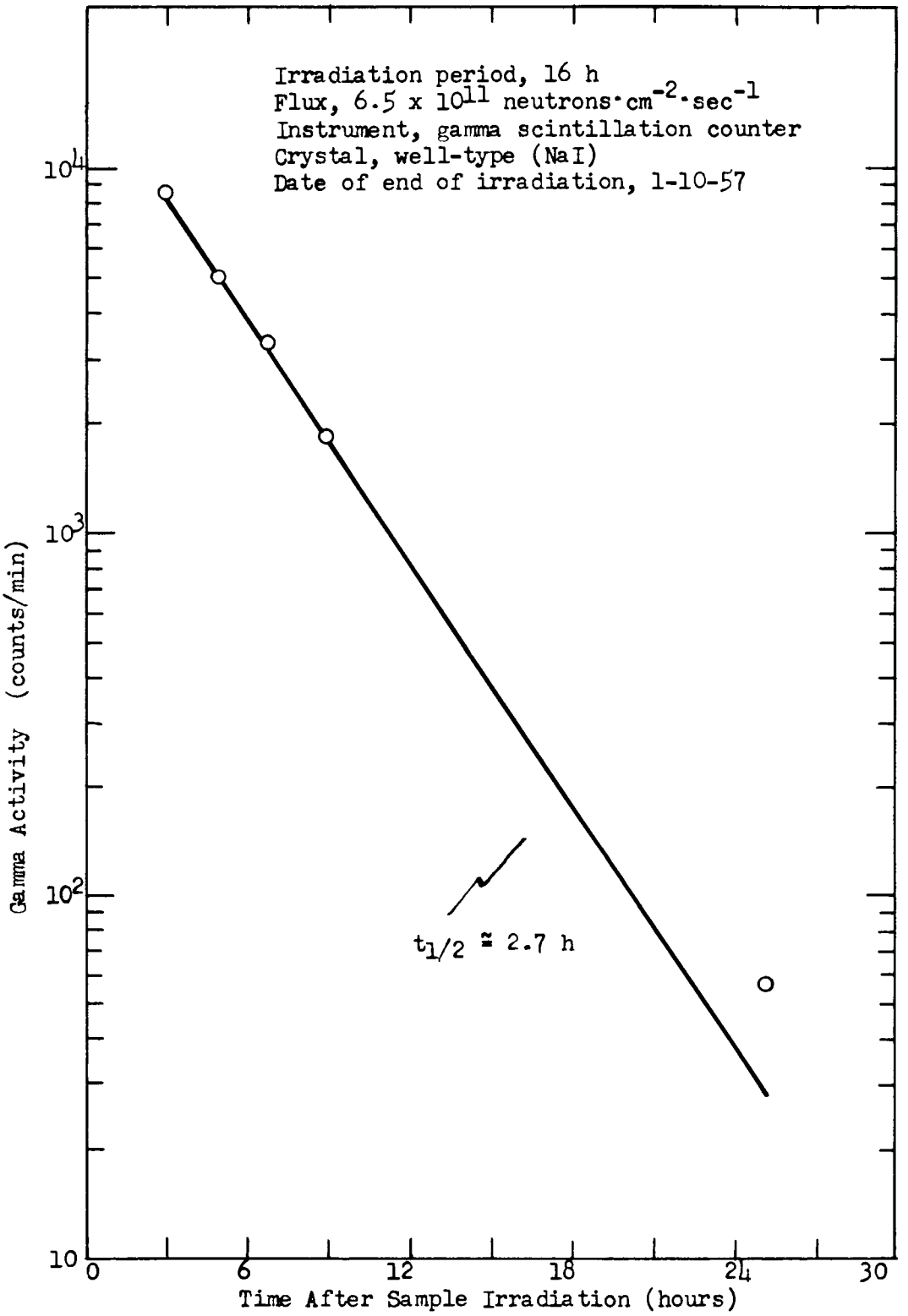

Fig. 63. Gross Gamma Decay for Neutron Irradiated Effluent from LITR Mixed-Bed Exchanger: Basic Sulfide Group.

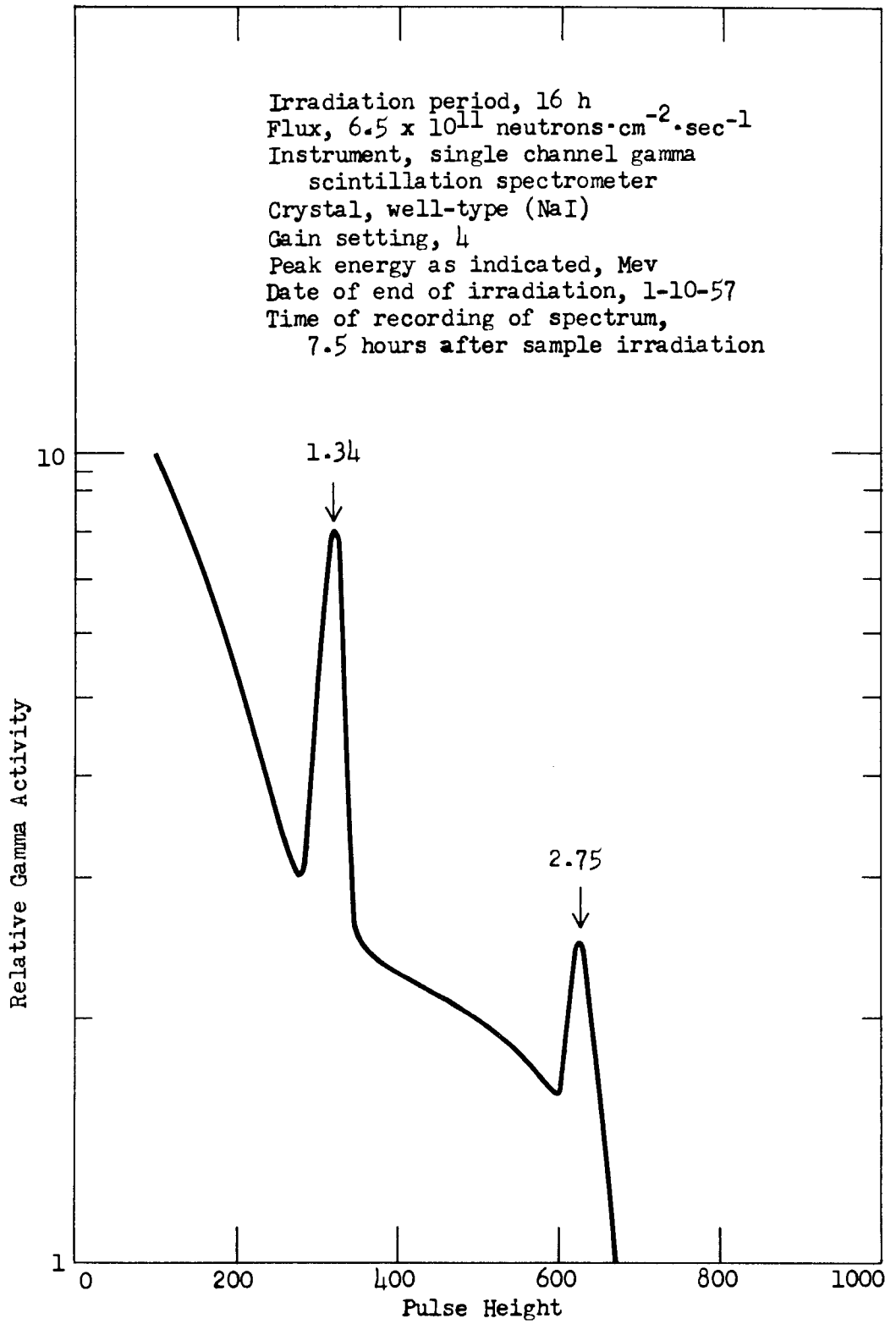

Fig. 64. Gamma Spectral Data for Neutron Irradiated Effluent from LITR Mixed-Bed Exchanger: Soluble Group.

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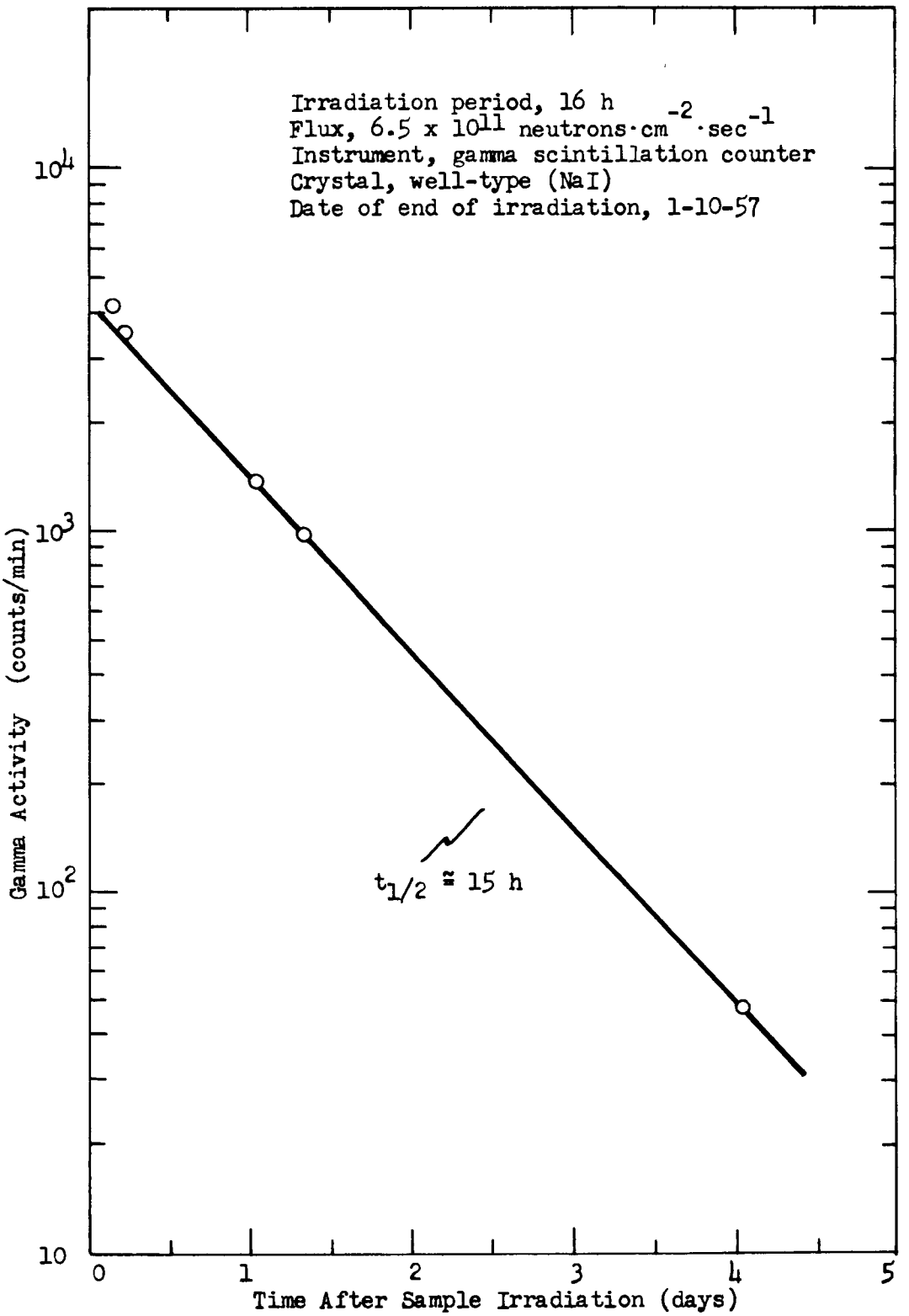

Fig. 65. Gross Gamma Decay for Neutron Irradiated Effluent from LITR Mixed-Bed Exchanger: Soluble Group.

TABLE VII

Concentrations of Stable Elements in Effluent from LITR Mixed-Bed Exchanger as Determined by Neutron Activation Analysis

Irradiation period, 16 h
Flux, 6.5×10^{11} neutrons·cm^{-2}·sec^{-1}
Date of sample collection, 12-17-56
Date of end of irradiation, 1-10-57

Chemical Group	Induced Radio-nuclide	Half Life	Estimated Concentration (μc/ml)*	Stable Element Source	Estimated Stable Element Concentration (ppm)
Silver	Br^{80m}	4.6 h	3×10^{-4} } 3×10^{-3}	Bromine	1×10^{-2}
	Br^{82}	35.9 h			
	Mn^{56}	2.58 h	5×10^{-4}	Manganese	2×10^{-4}
Acid Sulfide	Cu^{64}	12.8 h	3×10^{-5}	Copper	1×10^{-4}
Hydroxide and Alkaline Earth	F^{18}	1.87 h	2×10^{-4}	Oxygen	---
Basic Sulfide	Mn^{56}	2.58 h	7×10^{-4}	Manganese	3×10^{-4}
Soluble	Na^{24}	15.0 h	1×10^{-3}	Sodium	1×10^{-2}

* At end of irradiation.

when it is realized that both samples represent essentially a demineralized water. The similarity of the two samples affords an opportunity for evaluation of the single channel versus the 20 channel gamma scintillation spectrometer for routine water analysis. In general, spectral data were obtained more quickly and with greater definitiveness through use of the 20 channel unit.

G. Summary

The radioactive nuclides identified in the water at various stages of the LITR cooling water purification system are summarized in Table VIII.

The mode of decay and the more prominent characteristic gamma energies used in the identification of each of the radioactive nuclides in the cooling water are given in Table IX (Stehn, et al., 1956; Heath, 1956; Moore and Reynolds, 1957; Hollander, et al., 1953).

TABLE VIII

Summary of Radioactive Nuclides Identified in Water at Various Stages of LITR Cooling Water Purification System

Estimated concentrations given in units of 10^{-5} μc/ml

Make-Up Water		Cation Exchanger Influent		Cation Exchanger Effluent		Anion Exchanger Effluent		Mixed-Bed Exchanger Effluent	
Radio-nuclide	Est. Conc.*	Radio-nuclide	Est. Conc.**	Radio-nuclide	Est. Conc.**	Radio-nuclide	Est. Conc.**	Radio-nuclide	Est. Conc.**
Na^{24}	500	I^{131}, I^{132}, I^{133}, I^{135}	300	I^{131}, I^{132}, I^{133}, I^{135}	200	I^{132}, I^{133}	3	Cu^{64}	1
Br^{80}, Br^{80m}, Br^{82}	80	Na^{24}	300	Tc^{99m}	50	Cu^{64}	1	Ce^{143} + Pr^{143}	0.4
F^{18}	30	Cu^{64}	100	F^{18}	5	Te^{132} + I^{132}	1	Mo^{99} + Tc^{99m}	0.3
Mn^{56}	10	Np^{239}	80	Sb^{122}	2	Mo^{99} + Tc^{99m}	0.3	Te^{132} + I^{132}	0.2
Cu^{64}	4	Tc^{99m}	70	Pa^{233}	1	Ce^{143} + Pr^{143}	0.3	Ce^{141}	0.1
		Mn^{56}	70			Sb^{122}	0.2	Cr^{51}	0.1
		Sr^{91} + Y^{91m}	50			Ce^{141}	0.1	I^{131}, I^{132}, I^{133}, I^{135}	<0.01
		Pa^{233}	1			Cr^{51}	0.1		
		Co^{60}	1			Fe^{59}	0.05		
		Ba^{140} + La^{140}	1			I^{131}	0.03		

* At end of 16 hour irradiation at flux of 6.5×10^{11} neutrons·cm^{-2}·sec^{-1}.

** At time of sample collection.

TABLE IX

Radiation Characteristics of Radionuclides Identified in LITR Cooling Water

Radio-nuclide	Half Life	Mode of Decay	Prominent Gamma Energies (Mev)	Other Gamma Energies (Mev)
Ba^{140}	12.8 d	β^-, γ	0.54, 0.31, 0.16	
Br^{80}	18 m	$K, \beta^-, \beta^+, \gamma$	0.62	
Br^{80m}	4.6 h	IT, γ	0.04	
Br^{82}	35.9 h	β^-, γ	0.55, 0.77, 1.04	0.6 - 2
Ce^{141}	32 d	β^-, γ	0.145	
Ce^{143}	33 h	β^-, γ	0.29, 0.66, 0.72	0.06 - 1.10
Co^{60}	5.2 y	β^-, γ	1.33, 1.17	
Cr^{51}	27 d	K, γ	0.32	
Cu^{64}	12.8 h	$K, \beta^-, \beta^+, \gamma$	1.34	
F^{18}	1.87 h	β^+		
Fe^{59}	45 d	β^-, γ	1.10, 1.29, 0.19	
I^{131}	8.05 d	β^-, γ	0.36, 0.64	0.08 - 0.72
I^{132}	2.3 h	β^-, γ	0.67, 0.78	0.53 - 2.2
I^{133}	21 h	β^-, γ	0.53, 0.85, 1.4	
I^{135}	6.7 h	β^-, γ	1.3, 1.8, 2.4	0.53, 0.84, 1.13
La^{140}	40.2 h	β^-, γ	1.60, 0.82, 0.49	0.11 - 2.9
Mn^{56}	2.58 h	β^-, γ	0.85, 1.8	2.1 - 3
Mo^{99}	67 h	β^-, γ	0.74	0.04 - 0.78
Na^{24}	15.0 h	β^-, γ	2.75, 1.37	

TABLE IX (continued)

Radiation Characteristics of Radionuclides Identified in LITR Cooling Water

Radio-nuclide	Half Life	Mode of Decay	Prominent Gamma Energies (Mev)	Other Gamma Energies (Mev)
Np^{239}	2.33 d	β^-,γ	0.09, 0.21, 0.28	0.045 - 0.49
Pa^{233}	27.4 d	β^-,γ	0.09, 0.31, 0.075	0.016 - 0.42
Sb^{122}	2.8 d	β^-,K,β^+,γ	0.56	0.7 - 1.2
Sr^{91}	9.7 h	β^-,γ		0.65 - 1.41
Sr^{92}	2.7 h	β^-,γ	1.38	
Tc^{99m}	6.0 h	IT,γ	0.14	
Te^{132}	77 h	β^-,γ	0.23	
Xe^{131m1}	4.8×10^{-10} s	IT	0.08	
Xe^{133} + Cs^{133m}	5.27 d	β^-,γ	0.08	
Xe^{135} + Cs^{135m}	9.2 h	β^-,γ	0.25, 0.62, 0.37	
Y^{91m}	50 m	IT	0.55	

CHAPTER VII

DISSOLVED GAS ANALYSIS

Early studies of water samples from the LITR cooling system revealed that a sizeable portion of the total activity was readily removed by aeration. For example, over 95% of the gross gamma activity in the effluent from the mixed-bed exchanger was due to dissolved radioactive gases. As a result, it was deemed important that the identity of these dissolved gases be established.

For the gas analysis the following procedure was used. A sample of the effluent from the mixed-bed exchanger was collected, taking care to retain as much of the dissolved gas activity as possible. A 5 milliliter aliquot of this sample was then sealed within a small glass vial which would fit into the well of the gamma scintillation crystal, provided the aluminum absorber were removed. (This procedure was adopted to allow analysis of a larger volume sample.) An aerated 5 milliliter aliquot of the sample was sealed within a similar glass vial. Gamma spectral studies were then made on both the aerated and non-aerated aliquots using the 20 channel scintillation spectrometer. Gamma spectral data for the dissolved radioactive gases were obtained by subtracting the individual channel readings for the aerated sample from those for the non-aerated sample. A plot of these data is shown in Figure 66.

Two radioactive gases predominate. These are 1.82 h A^{41} shown by the prominent 1.31 Mev gamma peak and 9.2 h Xe^{135} characterized by the 0.25 Mev peak. Other gases in evidence, but

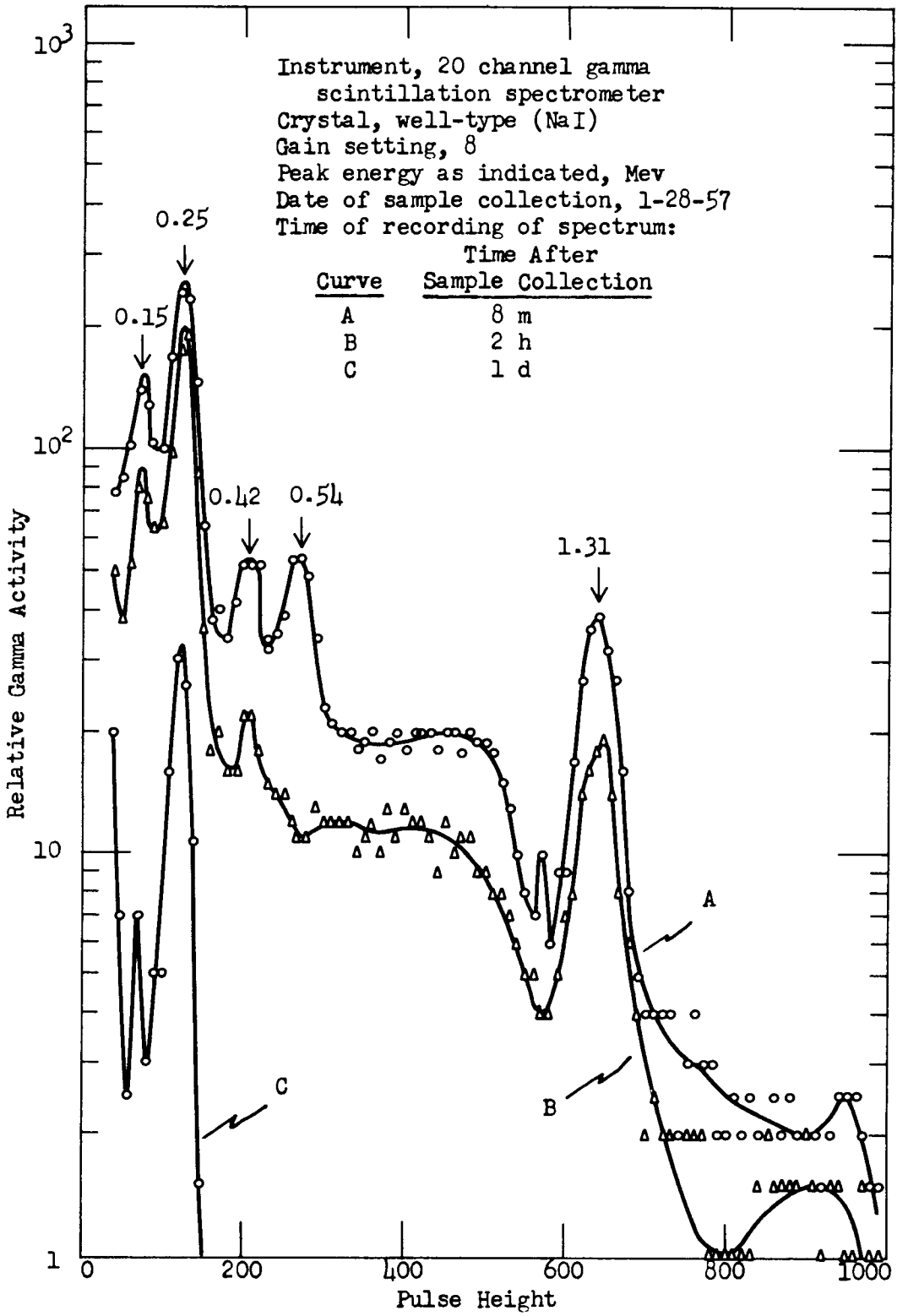

Fig. 66. Gamma Spectral Data for Effluent from LITR Mixed-Bed Exchanger: Radioactive Gas Nuclides.

in smaller concentration, include 15 m Xe^{135m} associated with the short-lived 0.54 Mev gamma peak and 78 m Kr^{87} associated with the 0.42 Mev peak. The 0.15 Mev peak probably represents 4.4 h Kr^{85m}.

Gamma spectral data at the lower energies are shown in Figure 67. These data show evidence for 2.8 h Kr^{88} and 5.27 d Xe^{133} as characterized by the 0.028 and 0.086 Mev peaks, respectively.

Several of the identified radioactive gas nuclides were known to be of fission product origin. However, some of these, namely Kr^{87}, and all the xenon isotopes, could also result from neutron irradiation of stable isotopes of the same elements. As a check on possible radioactive gas nuclides which might be induced, a sample of LITR cooling system make-up water was irradiated in the Hole 10 facility of the ORNL Graphite Reactor for 16 hours. Again aerated and non-aerated aliquots were prepared and the gamma spectrum for the gas activities was determined. The resulting data are shown in Figure 68. The predominant induced activity was 1.82 h A^{41}. The small gamma peak at approximately 0.23 Mev may be due to 9.2 h Xe^{135}. Spectral data for the 0.02 to 0.10 Mev range revealed no significant gamma peaks which could be attributed to induced activities resulting from neutron irradiation of stable isotopes of krypton and xenon.

Table X contains a summary of the radioactive gas nuclides identified in the cooling water and their probable origin. The total concentration of gas activity in the mixed-bed effluent at the time of sampling was approximately 3×10^{-3} μc/ml.

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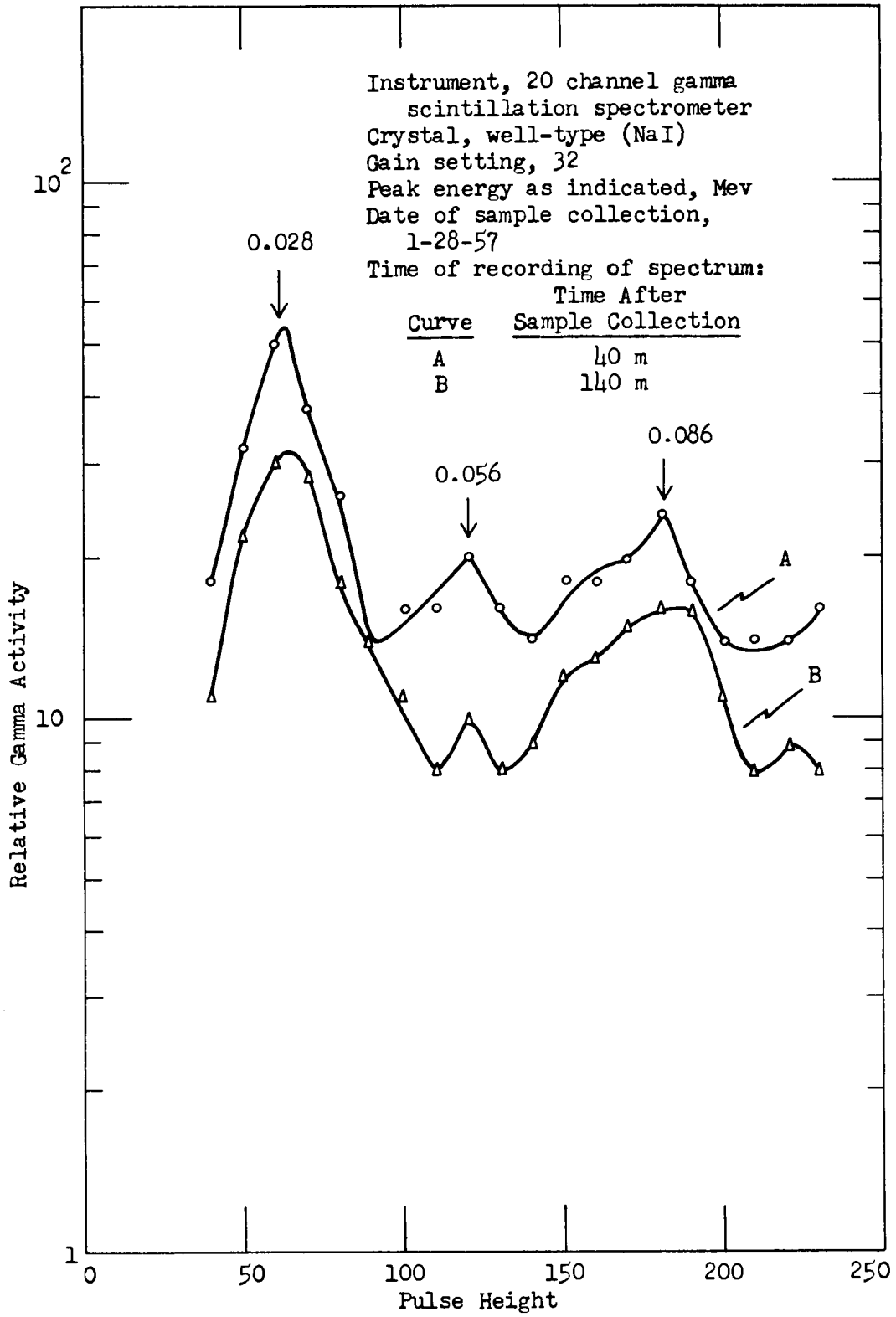

Fig. 67. Gamma Spectral Data for Effluent from LITR Mixed-Bed Exchanger: Radioactive Gas Nuclides.

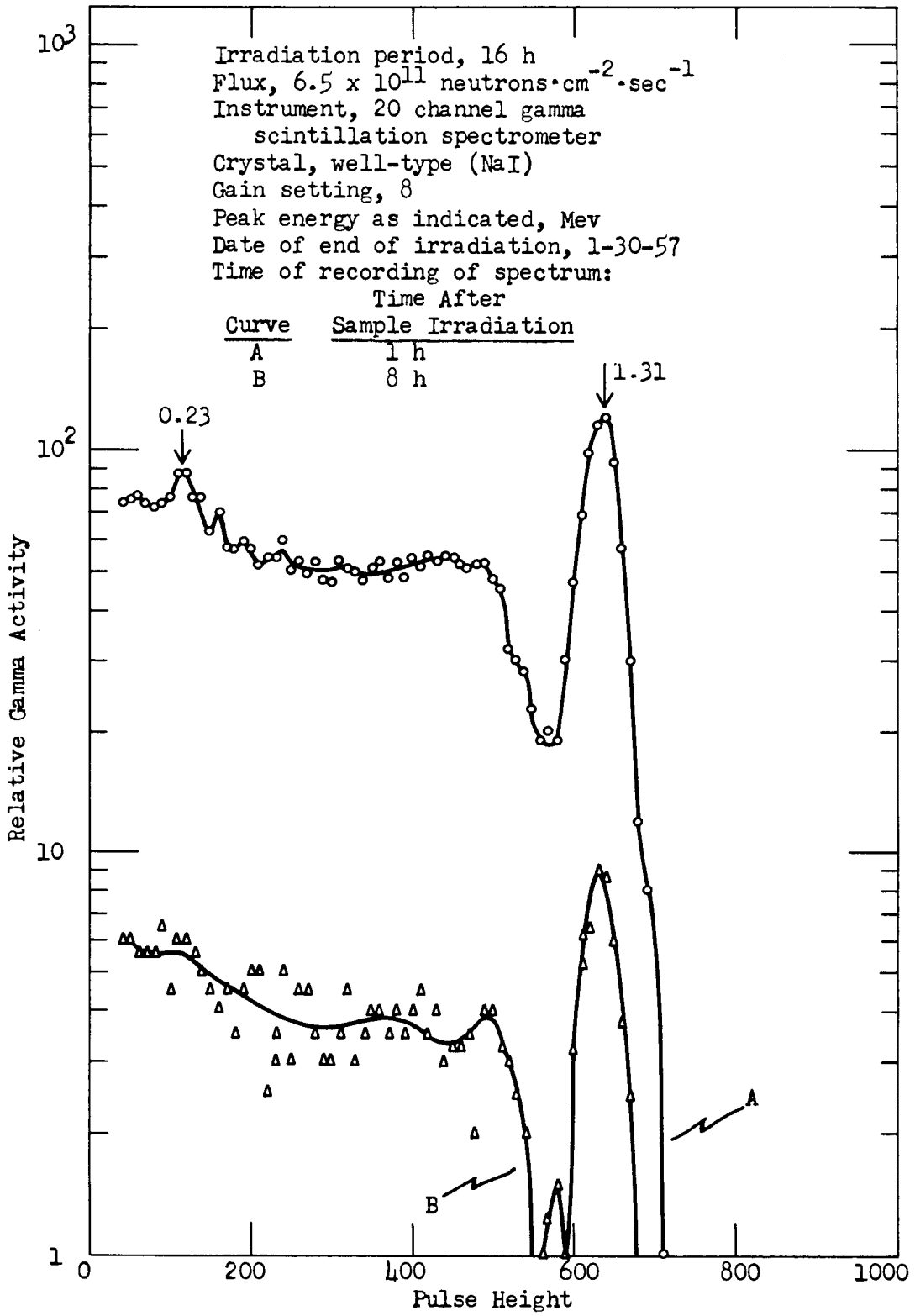

Fig. 68. Gamma Spectral Data for Neutron Irradiated LITR Make-Up Water: Radioactive Gas Nuclides.

TABLE X

Radioactive Gas Nuclides Identified in Effluent from the LITR Mixed-Bed Exchanger

Date of sample collection, 1-28-57

Radioactive Nuclide	Half Life	Probable Mode of Formation
A^{41}	1.82 h	$A^{40}(n,\gamma)A^{41}$
Xe^{135}	9.2 h	Fission Product
Xe^{135m}	15 m	Fission Product
Xe^{133}	5.27 d	Fission Product
Kr^{85m}	4.4 h	Fission Product
Kr^{87}	78 m	Fission Product
Kr^{88}	2.8 h	Fission Product

Note: The total concentration of dissolved gas activity in the sample at time of collection was approximately 3×10^{-3} μc/ml.

CHAPTER VIII

ION EXCHANGE RESIN STUDIES

As an additional study of the cooling water system, samples of the resin from each of the three ion exchange columns were collected and analyzed.

A. Gross Gamma Activity as a Function of Depth

The initial study consisted of a determination of the gross gamma activity in the columns as a function of depth. Samples of the cation and mixed-bed resins were collected on December 28, 1956. An attempt to sample the anion column on this same date was unsuccessful. A valve, mistakenly left open, disrupted the bed making analysis of this sample at this time impossible. As a result, the larger anion resin column was put into service. This column was then sampled on January 14, 1957.

Samples were collected at depth intervals of six inches, beginning at the surface of each bed and continuing to the bottom. At the time of sampling, the resin in each of the beds had settled for several hours.

For gross gamma determinations samples of the resins were transferred to 10 x 75 millimeter cork stoppered Pyrex culture tubes and counted in the well-type scintillation counter. Counts were made approximately 24 hours after collection of the samples. Later the samples were dried and weighed in order that the gamma

activities could be recorded as functions of dry resin weights. The data are shown in Figures 69, 70 and 71. It will be noted that for all three resin columns, the major portion of the gross gamma activity is in the top layer of the bed.

B. Apparent Half Life as a Function of Depth

Gross gamma decay determinations comprised the second portion of the resin study. Samples used for these determinations were similar to those prepared for the gross gamma activity study previously described.

Decay data for resin samples collected at several depth intervals within each of the three ion exchange columns are shown in Figures 72, 73 and 74. In general, the over-all half life of the radioactive material retained on the resin increased with column depth.

C. Radioactive Nuclides

The third portion of the resin study was directed toward identification of the major radionuclides present on the ion exchange columns at the surface and bottom bed levels. Data as to the composition of the radioactive materials at these depths were deemed important for several reasons: (a) there should be close correlation between the radionuclides in the upper levels of the bed and those found removed from the water in its passage through

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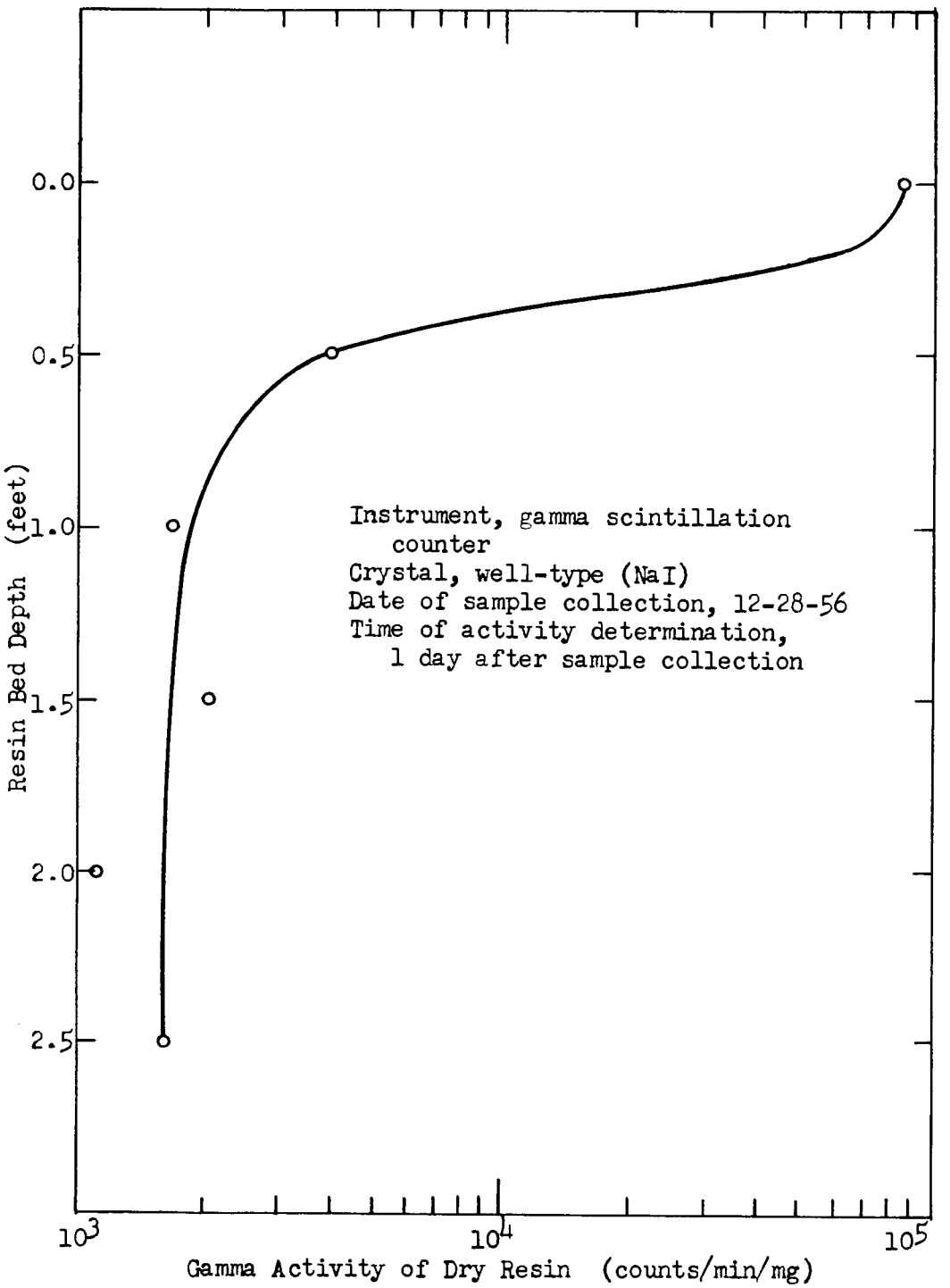

Fig. 69. Gross Gamma Activity as Function of Resin Bed Depth: LITR Cation Exchanger.

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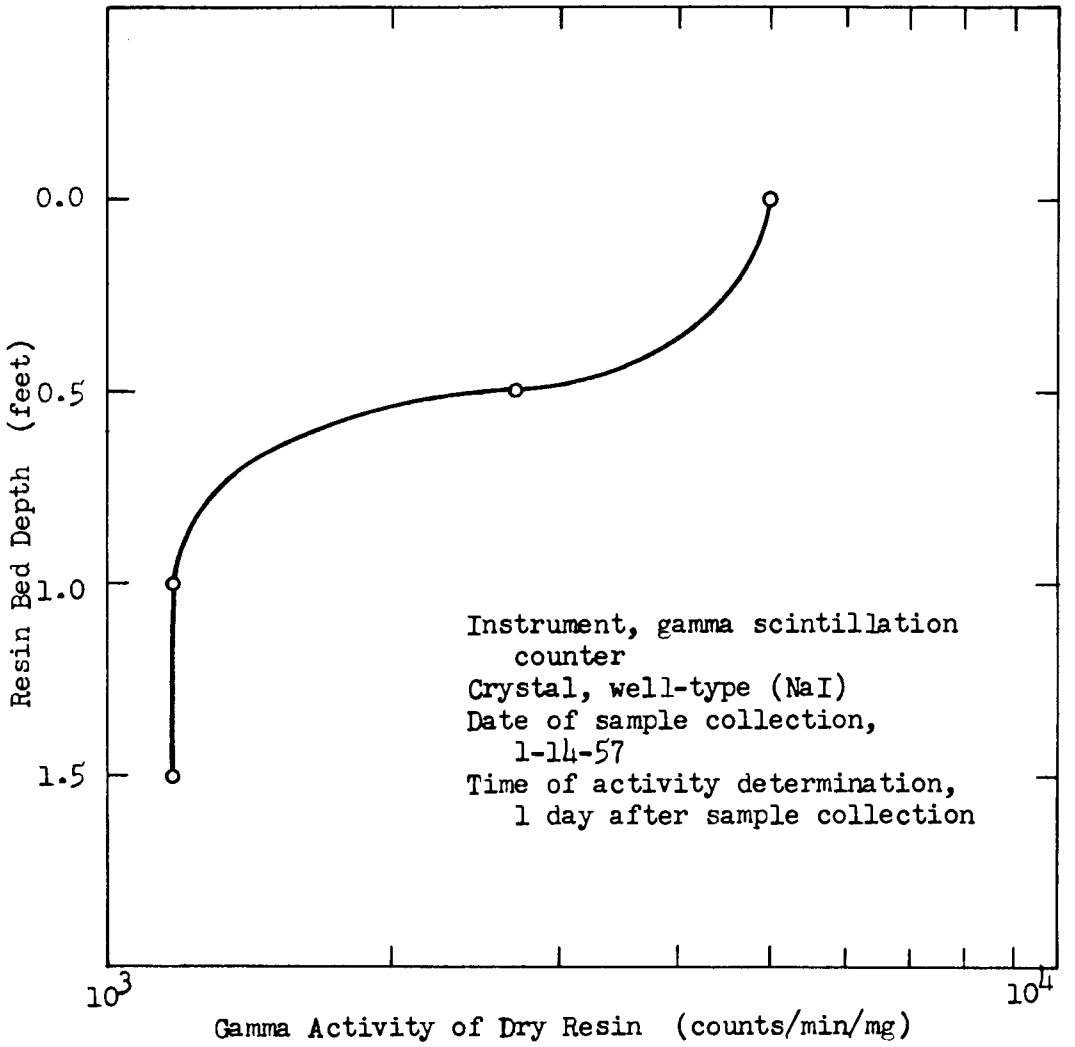

Fig. 70. Gross Gamma Activity as Function of Resin Bed Depth: LITR Anion Exchanger.

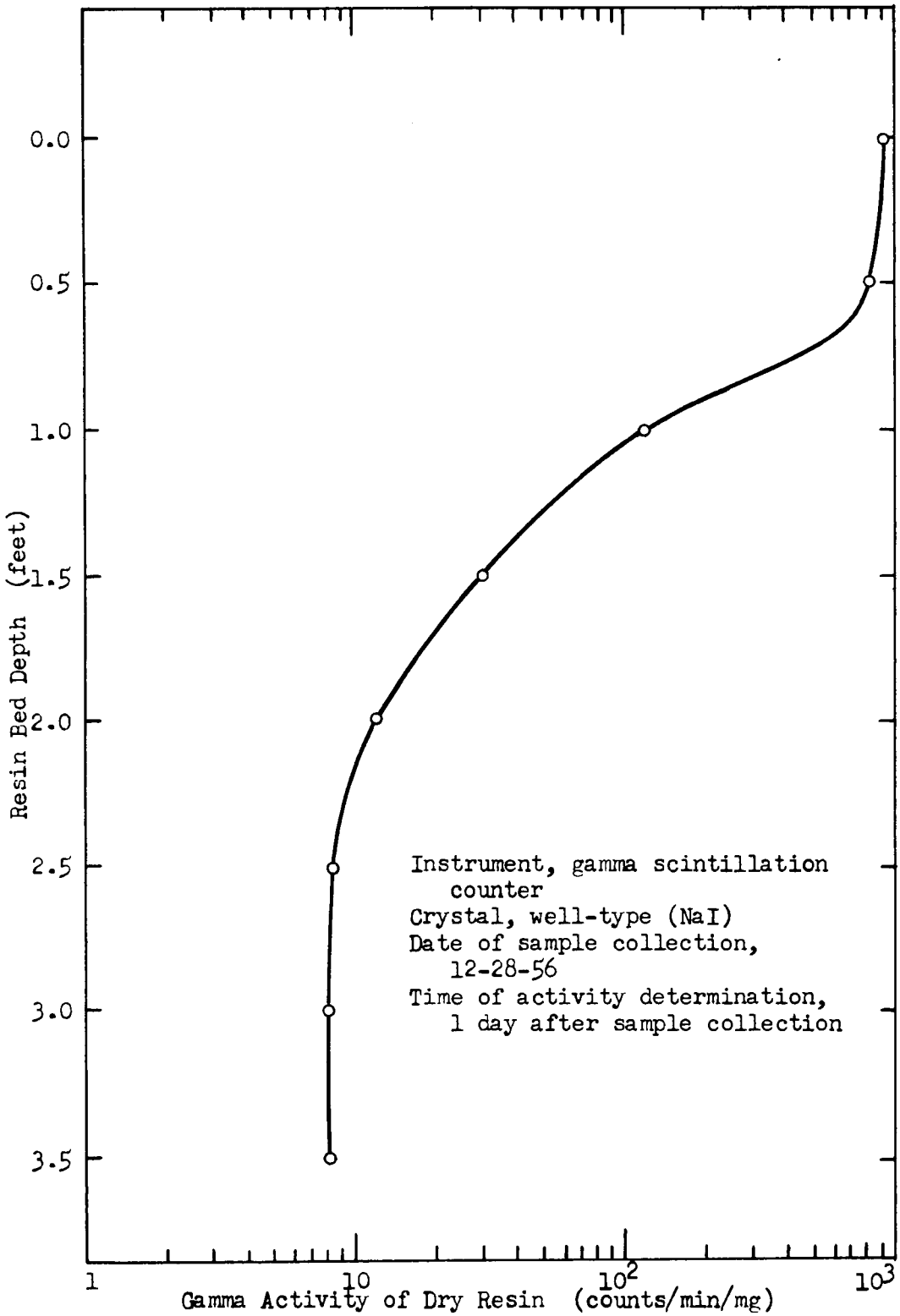

Fig. 71. Gross Gamma Activity as Function of Resin Bed Depth: LITR Mixed-Bed Exchanger.

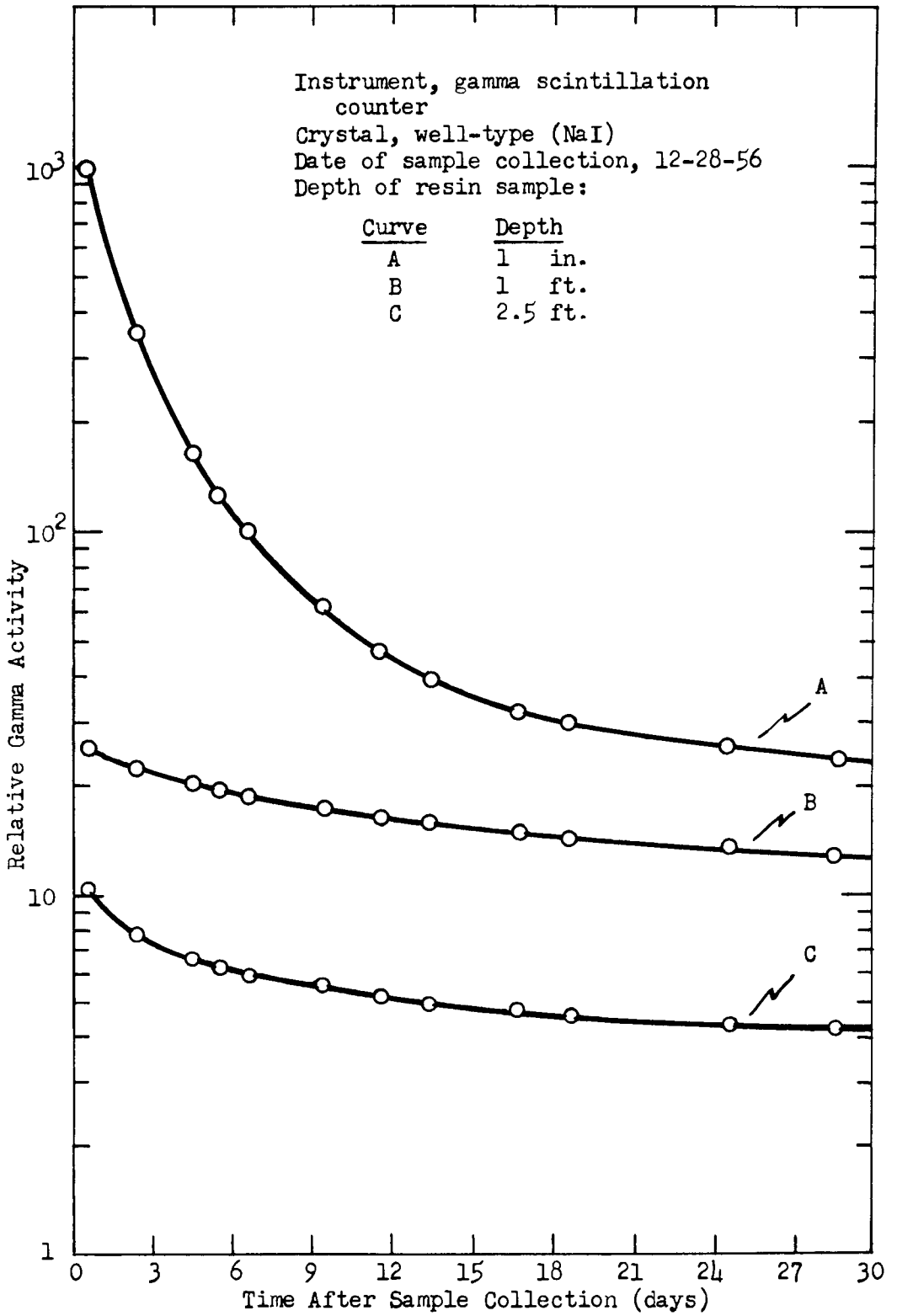

Fig. 72. Gross Gamma Decay as Function of Resin Bed Depth: LITR Cation Exchanger.

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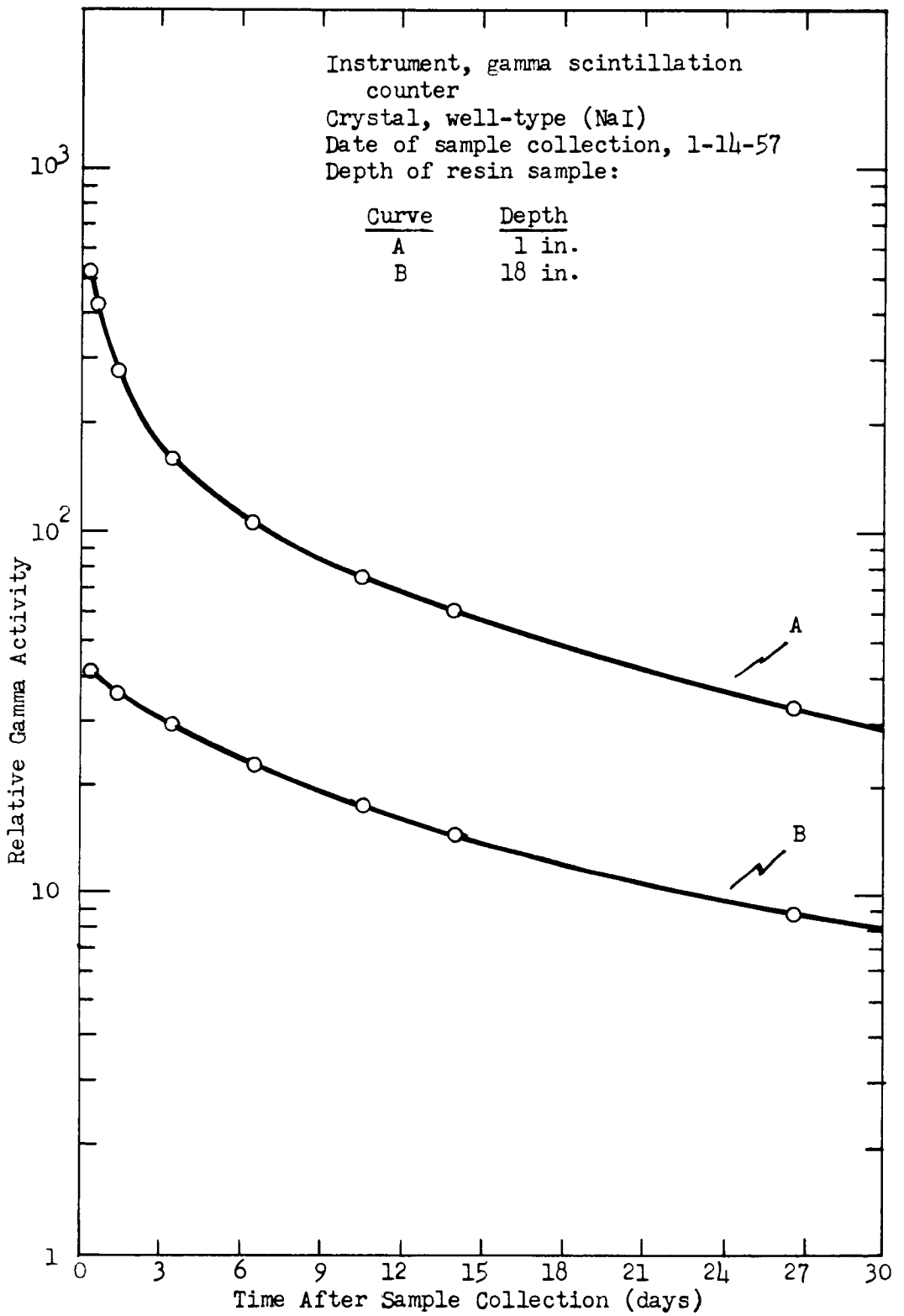

Fig. 73. Gross Gamma Decay as Function of Resin Bed Depth: LITR Anion Exchanger.

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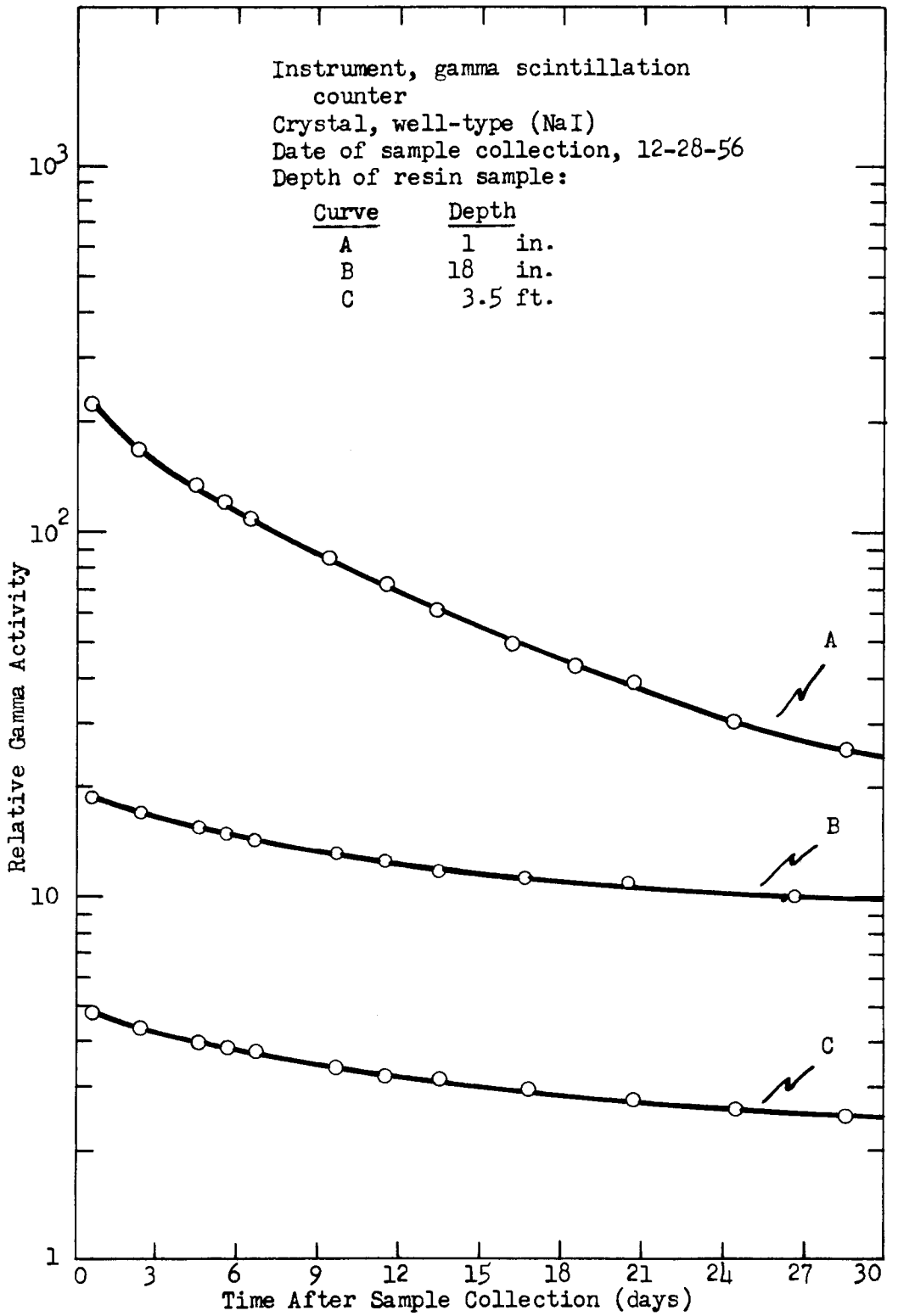

Fig. 74. Gross Gamma Decay as Function of Resin Bed Depth: LITR Mixed-Bed Exchanger.

a particular column, (b) radionuclides found at the bottom of the bed should perhaps be somewhat poorly removed from the water, their presence at this level indicating near saturation of the column with that particular radioactive nuclide.

Inasmuch as the objective of this particular study was to determine the major radionuclides only, gamma spectral determinations were made on the resin samples as removed from the column, no radiochemical separation techniques being employed.

1. Cation Resin

Gamma spectral data for a sample of surface level cation resin are shown in Figures 75 and 76. The predominant radionuclide the first few days after sampling was 15.0 h Na^{24} as characterized by the 1.34 and 2.75 Mev gamma peaks of Figure 75. After decay of Na^{24} the predominant activity is due to 2.33 d Np^{239} shown by the 0.10 and 0.21 Mev gamma peaks of Figure 76. With further decay 245 d Zn^{65} appears. This nuclide is characterized by the gamma peak at 1.10 Mev. The long-lived 1.34 Mev peak represents 5.2 y Co^{60}, its 1.17 Mev gamma peak being overshadowed by the 1.10 Mev peak of Zn^{65}. The 0.47, 0.80 and 1.59 Mev peaks probably represent 40.2 h La^{140}, the daughter of 12.8 d Ba^{140}.

Gamma spectral data for a resin sample from the bottom of the cation exchanger are shown in Figure 77. Here, as in the surface level sample, the presence of Co^{60} and Zn^{65} is indicated by the characteristic long-lived 1.34 and 1.11 Mev peaks, respectively. Again it appears that the characteristic 0.50, 0.83 and 1.59 Mev peaks of La^{140} are present.

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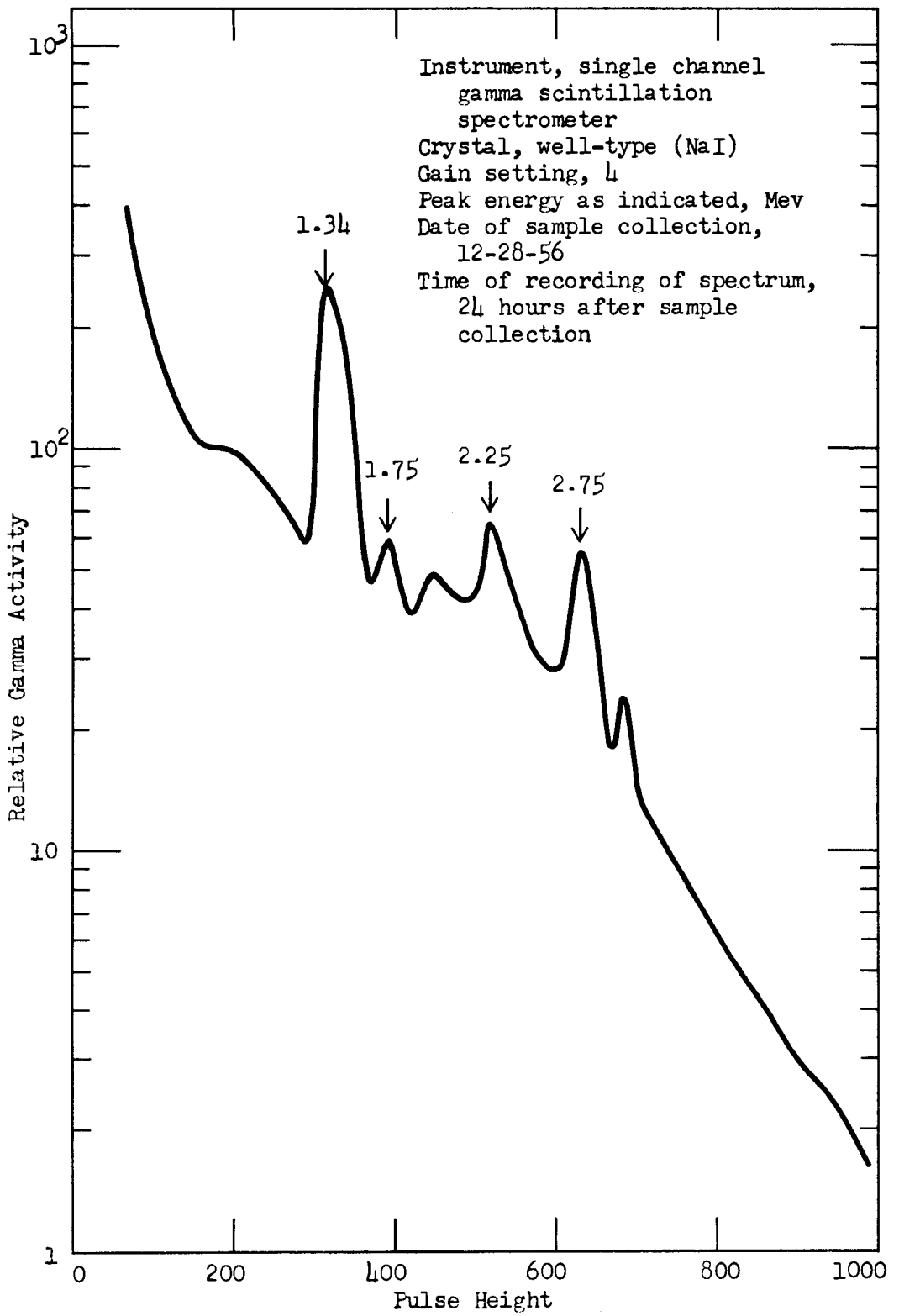

Fig. 75. Gamma Spectral Data for Surface Level Resin Sample: LITR Cation Exchanger.

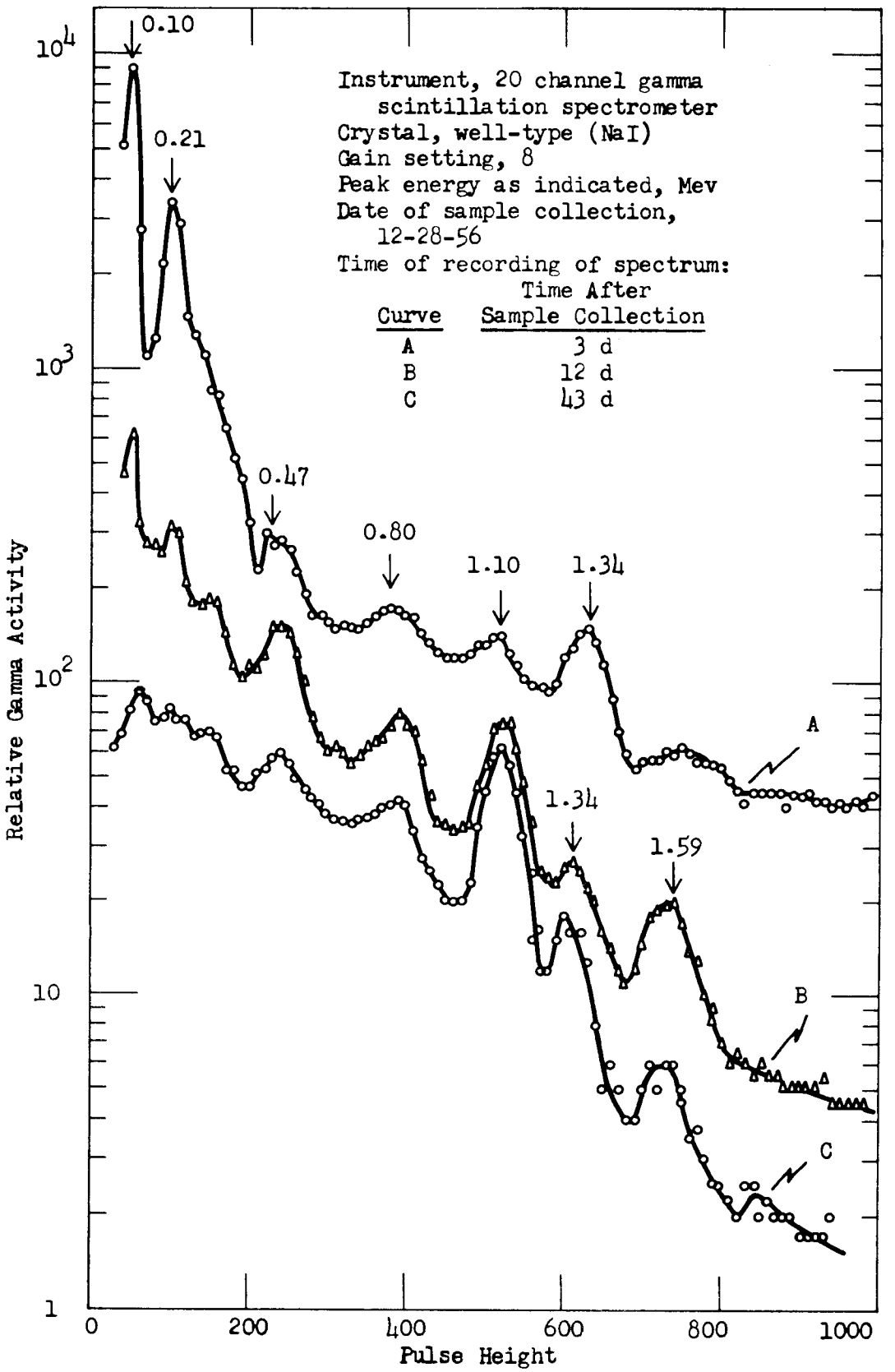

Fig. 76. Gamma Spectral Data for Surface Level Resin Sample: LITR Cation Exchanger.

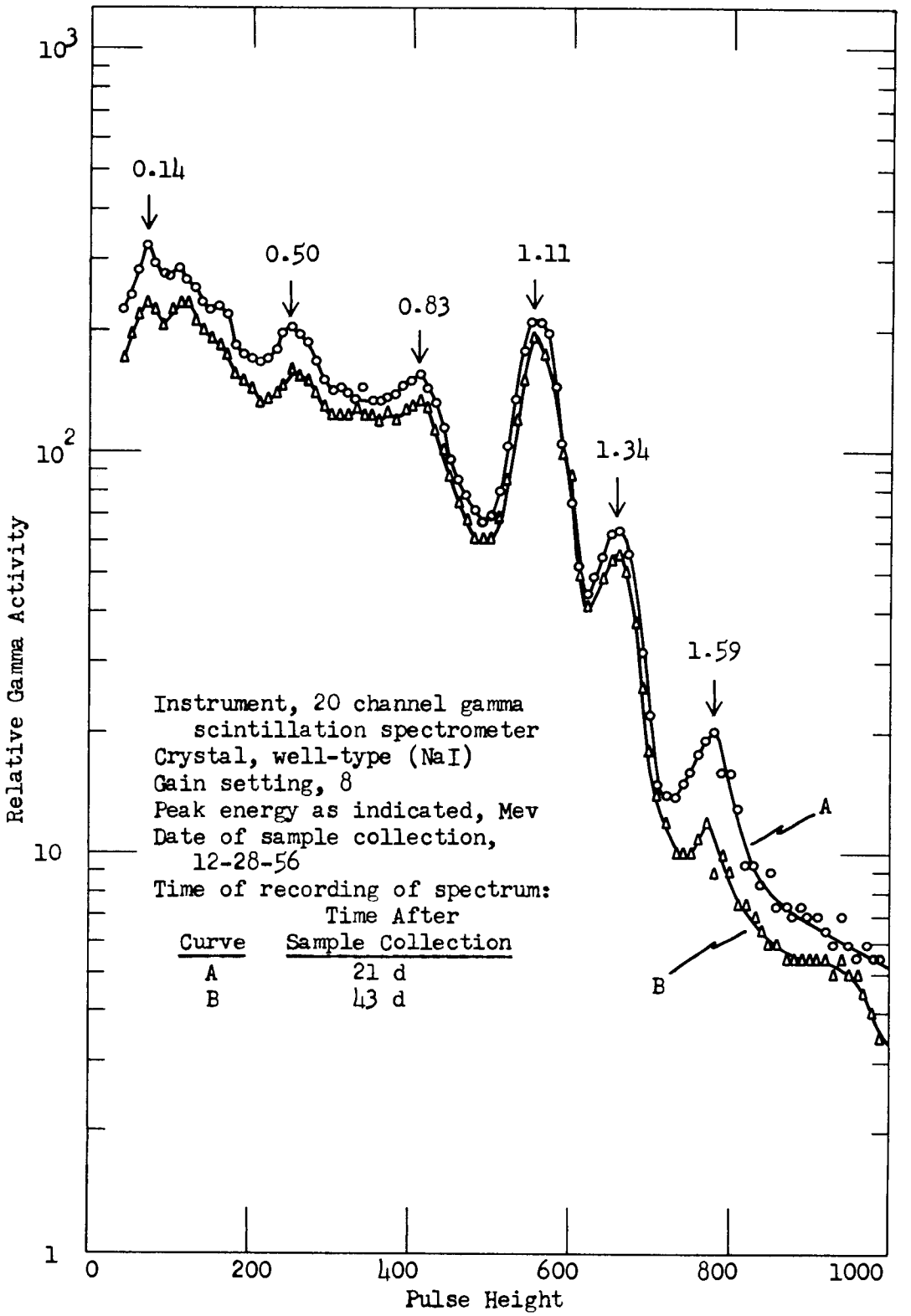

Fig. 77. Gamma Spectral Data for Bottom Level Resin Sample: LITR Cation Exchanger.

2. Anion Resin

Gamma spectral data for a surface level resin sample from the anion column are shown in Figure 78. The predominant radioactive nuclides initially present are 67 h Mo^{99} and its daughter 6.0 h Tc^{99m} as characterized by the 0.14 Mev gamma peak, 8.05 d I^{131} as shown by the 0.37 Mev peak and 21 h I^{133} as shown by the 0.54 Mev peak. Also probably present in lesser concentration is 77 h Te^{132} plus its daughter 2.3 h I^{132} as shown by the small 0.24 and 0.68 Mev gamma peaks. After several weeks decay I^{131} remains the principal activity.

Gamma spectral data for a bottom level resin sample from the anion column are shown in Figure 79. The predominant radioactive nuclide initially present was 8.05 d I^{131} as characterized by the 0.368 Mev gamma peak. The presence of 67 h Mo^{99} + 6.0 h Tc^{99m} is shown by the 0.143 Mev gamma peak. Spectral determinations at higher energies failed to reveal the presence of 21 h I^{133} as a major activity in this sample. It is quite possible that 27 d Cr^{51} was also present since it has a characteristic gamma peak at 0.32 Mev.

3. Mixed-Bed Resin

Gamma spectral and decay data for a surface level sample from the mixed-bed column are shown in Figure 80. Radioactive nuclides initially in predominance are 8.05 d I^{131} and 21 h I^{133} as characterized by the 0.37 and 0.55 Mev gamma peaks. The presence of 67 h Mo^{99} + 6.0 h Tc^{99m} is shown by the 0.14 Mev peak. The sources of the 0.58 and 1.69 Mev peaks were not identified.

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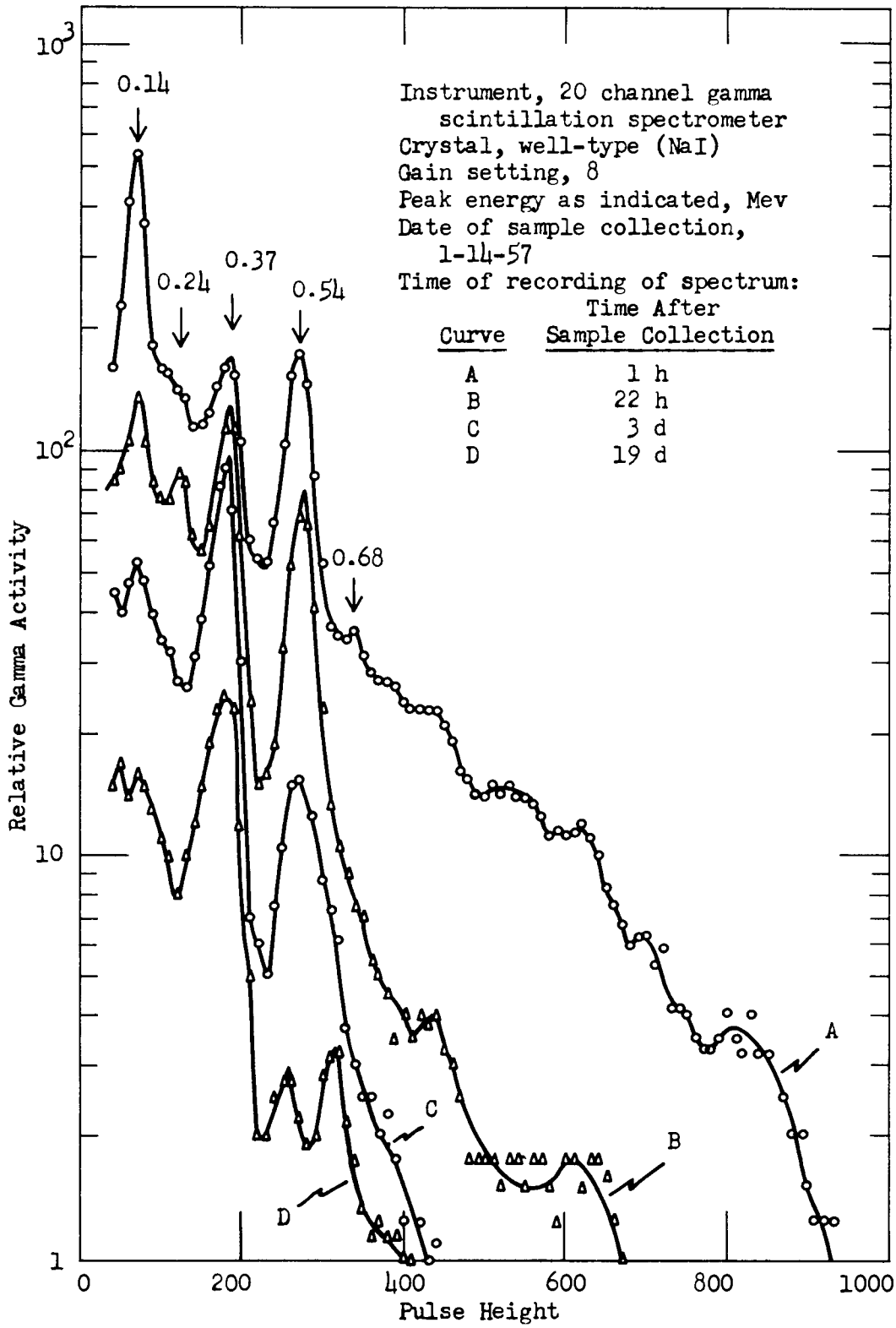

Fig. 78. Gamma Spectral Data for Surface Level Resin Sample: LITR Anion Exchanger.

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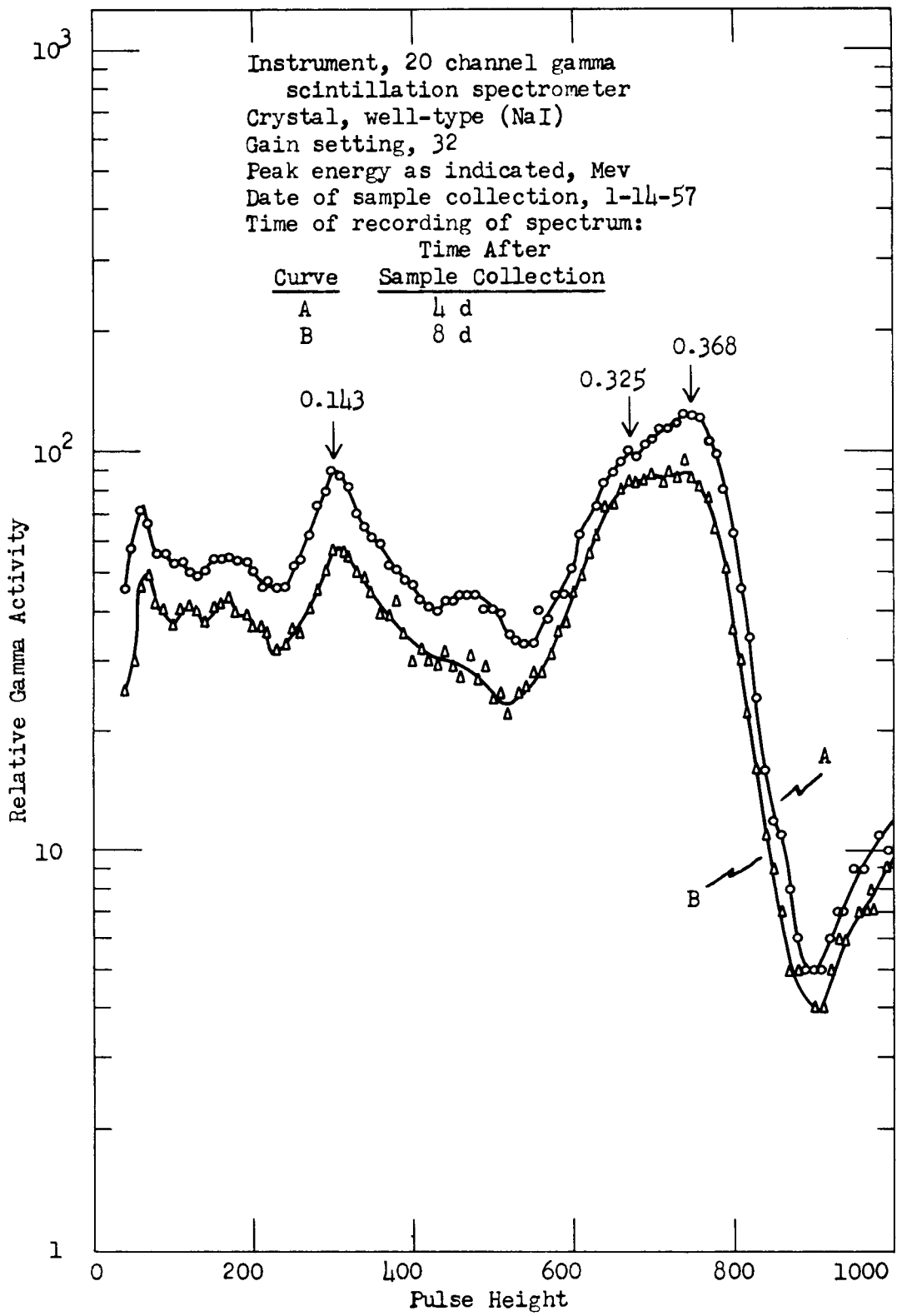

Fig. 79. Gamma Spectral Data for Bottom Level Resin Sample: LITR Anion Exchanger.

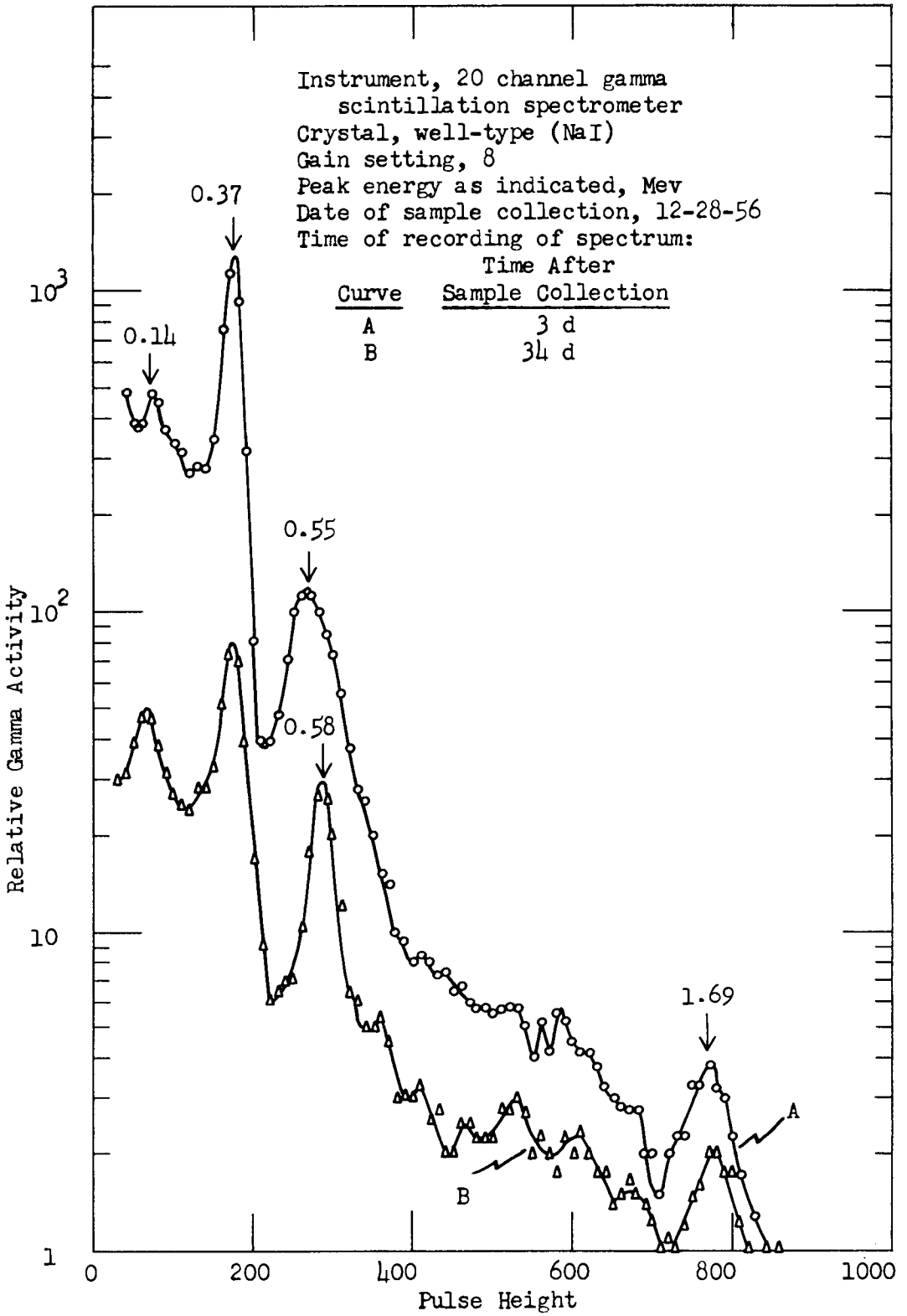

Fig. 80. Gamma Spectral Data for Surface Level Resin Sample: LITR Mixed-Bed Exchanger.

Gamma spectral and decay data for the bottom level resin sample from the mixed-bed column are shown in Figure 81. Radioactive nuclides in evidence are 32 d Ce^{141} as characterized by the 0.14 Mev peak and Co^{60} as shown by the 1.15 and 1.35 Mev peaks. The source of the 0.63 Mev gamma peak was not identified. It may be indicative of 30 y Cs^{137}. The source of the 1.55 Mev peak is also unknown.

4. Summary

A summary of the predominant radioactive nuclides identified on each of the three resin beds is given in Table XI.

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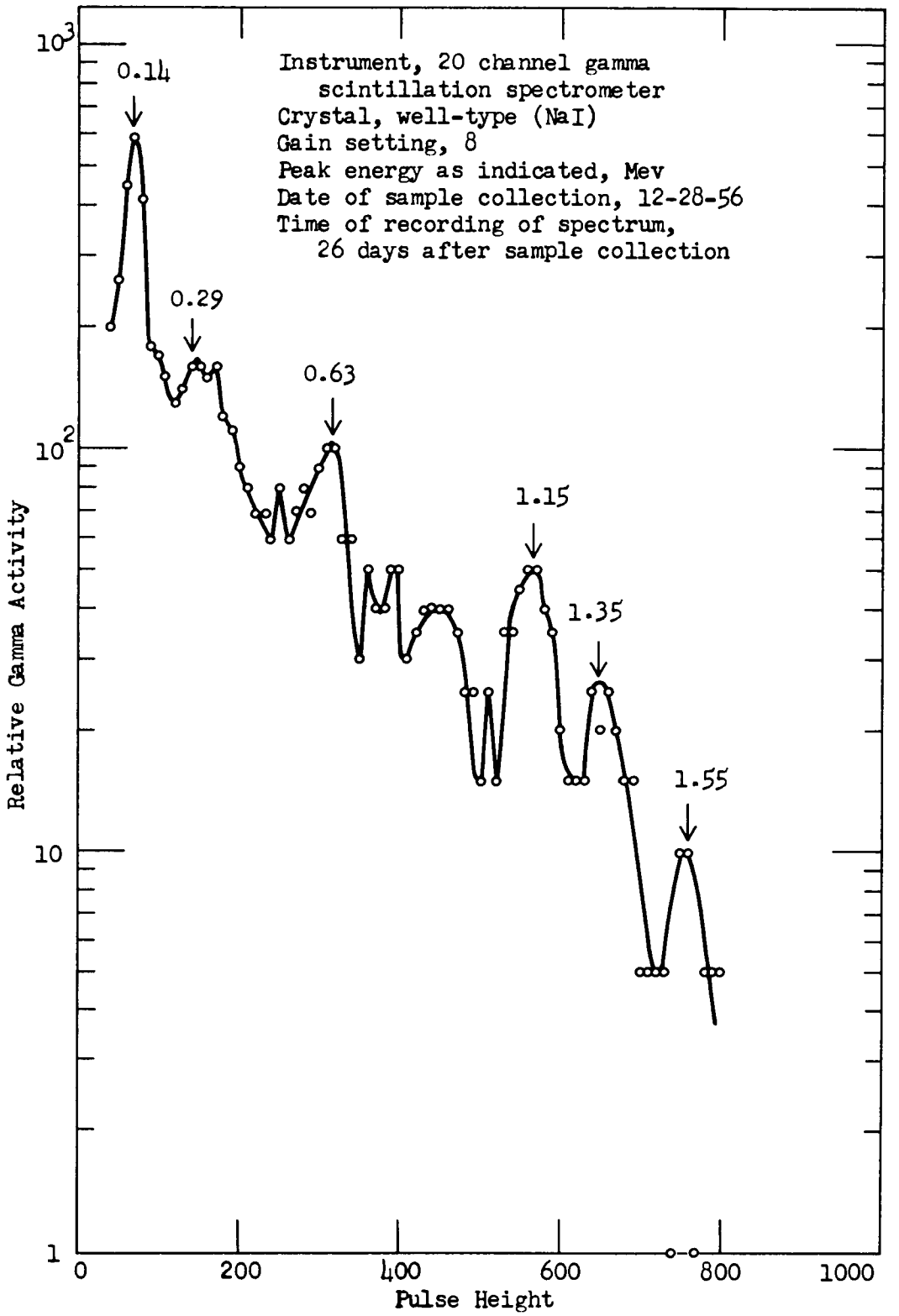

Fig. 81. Gamma Spectra Data for Bottom Level Resin Sample: LITR Mixed-Bed Exchanger.

TABLE XI

Radioactive Nuclides Identified on Ion Exchange Resins of LITR Cooling Water Purification System

Cation Resin		Anion Resin		Mixed-Bed Resin	
Radioactive Nuclide	Half Life	Radioactive Nuclide	Half Life	Radioactive Nuclide	Half Life
Na^{24}	15.0 h	I^{131}	8.05 d	I^{131}	8.05 d
Np^{239}	2.33 d	I^{133}	21 h	I^{133}	21 h
Zn^{65}	245 d	Mo^{99} + Tc^{99m}	67 h 6.0 h	Ce^{141}	32 d
Co^{60}	5.2 y	Te^{132*} + I^{132*}	77 h 2.3 h	Co^{60}	5.2 y
Ba^{140*} + La^{140*}	12.8 d 40.2 h	Cr^{51*}	27 d	Mo^{99*} + Tc^{99m*}	67 h 6.0 h

* Presence not confirmed.

CHAPTER IX

SOURCE OF RADIOACTIVE NUCLIDES

In order to complete the objectives of this study, it was desirable to establish the sources of the radioactive nuclides identified in the cooling water system. Where the sources of particular radionuclides could be established, better control of the release of these nuclides to the water system could perhaps be effected. The benefits of such control would include longer ion exchange resin life and lower radiation levels surrounding the cooling system proper.

Inasmuch as the sources of the dissolved radioactive gases identified in the cooling water have been reported elsewhere in this report (See Table X), no further discussion will be made here.

A. Neptunium and Protactinium

Perhaps the first radioactive nuclide whose source should be discussed is Np^{239}. A large quantity of this radionuclide would certainly not be expected in the cooling water of a reactor such as the LITR, where the fuel enrichment is in excess of 90% U^{235}, the most common method for the production of Np^{239} being:

$$U^{238}(n,\gamma)U^{239} \xrightarrow[23.5\ m]{\beta^-} Np^{239}.$$

The presence of Np^{239} in the cooling water of the LITR has been known for some time (Rupp and Cox, 1955; Lyon and Reynolds, 1955 b).

Quantitative data show that its concentration during the past three years has gradually decreased from about 3×10^{-1} μc/ml to 3×10^{-4} μc/ml, a reduction of approximately 10^3. During the same period the Na^{24} concentration in the water has decreased only by a factor of 10.

In view of the fact that known breaks occurred several years ago in natural uranium samples being exposed within the coolant stream of the reactor vessel, it is reasonable to assume that the major portion of the Np^{239} in the water has occurred as a result of natural uranium contamination within the reactor. The reduction in Np^{239} in the water would seem to indicate the gradual removal of this contamination from the system by the bypass purification units. The decrease in Na^{24} content is probably indicative of the improved performance of the cooling water ion exchange purification system during this same period.

The attribution of the source of Np^{239} to natural uranium contamination is substantiated by the fact that it has not been reported in the cooling water of the Materials Testing Reactor (Heath, 1956).

Protactinium probably originates from thorium contamination within the reactor vessel, there having been ample opportunity for such contamination during many tests conducted within the system in the past. Protactinium-233 decays with a half life of 27.4 days, and a common method for its production is:

$$Th^{232}(n,\gamma)Th^{233} \xrightarrow[23.3\ m]{\beta^-} Pa^{233}.$$

Protactinium-233 could also be produced by the reaction:

$$U^{235}(n,\gamma)U^{236}(n,\gamma)U^{237} \xrightarrow[6.75\ d]{\beta^-} Np^{237} \xrightarrow[2.2 \times 10^6\ y]{\alpha} Pa^{233}.$$

However, the extremely long half life of Np^{237} makes this source of considerably less importance.

B. Fission Products

The fission products make up the second group of radionuclides whose source should be discussed.

If the fission products identified in the water are tabulated, one interesting similarity is apparent. With the exception of Te^{132}, Mo^{99} + Tc^{99m} and the iodine group, they are all descendants of radioactive fission product gases. Since iodine and tellurium are among the more volatile fission products and since Mo^{99} + Tc^{99m} could be of induced as well as of fission product origin, it would appear that the main source of fission products in the water could be gaseous diffusion of the nuclides through the fuel element cladding.

Two isotopes of cesium are prominent fission products. One of these, 30 y Cs^{137}, is a descendant of the fission product gas Xe^{137}. The other isotope, 13 d Cs^{136}, however, is formed directly in fission. A check on the presence of these two isotopes in the cooling water system should furnish data either supporting or disproving the gaseous diffusion theory.

Cesium for the study was obtained by washing a surface level sample of cation resin with hot 6 N hydrochloric acid. The resin

sample was taken from the cation bed of the ion exchange purification system 51 days after the new column had been put into service. The sample was processed 18 days after removal from the bed, the cesium being separated from the other radioactive nuclides leached from the resin by precipitation of $CsClO_4$ from $HClO_4$ with absolute ethanol. The precipitate was then dissolved, several $Fe(OH)_3$ scavenge steps performed to remove contaminants, and the final cesium reprecipitated for the spectral studies (Hillebrand, et al., 1953).

Gamma spectral data for the separated cesium are shown in Figure 82. As in the other spectral studies reported herein, the well-type scintillation crystal was used. The prominent 0.66 Mev gamma peak represents Cs^{137}. The 0.16, 0.82, 1.02 and 1.38 Mev peaks represent Cs^{136}. Therefore, it must be concluded that the cooling water contained fission products directly yielded in fission as well as those descended from fission product gases.

Further study of the fission products identified in the water revealed a second similarity. In general, they are those of greatest fission yield. In addition, the presence in the water of the short-lived isotopes of iodine and the absence from the water of significant quantities of the high yield longer-lived gamma emitting fission products substantiated the fact that the fission products reach the water shortly after formation.

Based on this evidence, there are two possible sources for the fission products in the coolant:

(a) fission of natural or enriched uranium contamination within the reactor vessel,

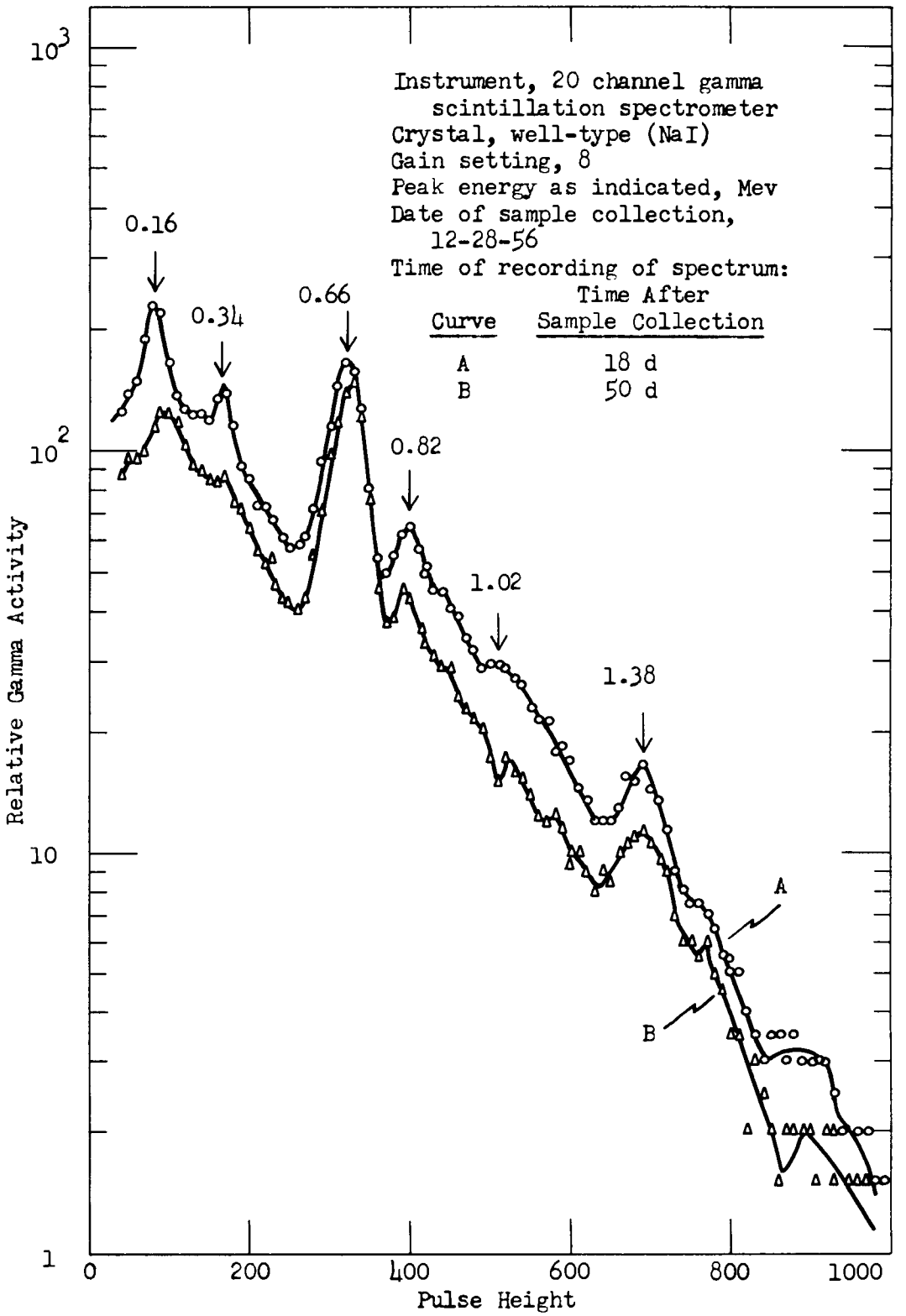

Fig. 82. Gamma Spectral Data for Surface Level Resin Sample from LITR Cation Exchanger: Cesium Separation.

(b) fission of enriched uranium in or on the fuel elements.

For either source, it would be assumed that the fission products enter the water by recoil at the time of fission.

If the reduction in the amount of Np^{239} in the water over the past several years is taken as evidence for the removal of natural uranium contamination from the system, and if the fission products have originated from this source, a comparable reduction in the quantity of fission products in the water should be in evidence. Quantitative determinations of the radioactive iodine in the water over the past several years show it has been reduced by a factor of 10 (Reynolds, 1957). This is the same amount by which the Na^{24} content has been reduced and probably again reflects only the improved performance of the ion exchange purification system. Comparison of quantitative data for neptunium, sodium and iodine would seem justified since all three radioactive elements were removed from the water by the ion exchange purification system with approximately equal effectiveness. Further, since their half lives are relatively short, data on these nuclides would tend to reflect conditions in the system near the time of sampling rather than at a time much earlier. Thus, the evidence does not point to natural uranium contamination as a major source of the fission products.

Enriched uranium contamination of the system should also be considered as a source of the fission products. However, in view of the knowledge that no enriched uranium samples have been exposed

within the coolant stream and no fuel element ruptures have occurred (with their possible release of U^{235} contamination into the system), this source seems unlikely.

The similarity in the reduction of radioactive sodium and iodine over the past several years tends to implicate the fuel elements themselves as the source of the fission products. For fission products to escape from the fuel elements to the water at the time of fission, the fissioning fuel would have to be at, near or on the surface of the element. At least three possible conditions could bring this about: (a) pitting of the fuel element cladding, (b) contamination of the fuel element cladding with enriched uranium during fabrication, (c) diffusion of the uranium into the cladding.

Visual examination of fuel elements exposed for a short time within the LITR has revealed the presence of small pits in the cladding surfaces. The observed pits were not thought, however, to be frequent or deep enough to permit escape of fission products. Due to the high levels of radiation involved, accurate data as to the size, depth and frequency of pits on fuel elements removed from the LITR after the normal 2 year cycle are not available. Estimation of conditions on such elements through extrapolation of the observed data for short-time exposure would be subject to serious errors.

During fabrication of the fuel elements, the enriched uranium-aluminum alloy is handled and worked in the same machinery where later the aluminum cladding is affixed to the fuel. Such a procedure could easily lead to contamination of the outer surfaces

of the cladding with the enriched fuel itself. Although the fuel plates are subjected to final acid cleansing, this procedure may not be adequate for removal of all possible contamination. Further, in the process of heat treatment following initial fabrication of the elements, there is the possibility for fuel to approach the cladding surface through diffusion. Of these two possibilities, it would appear that enriched uranium contamination of the cladding during fabrication would be the more probable.

To check on these latter two possibilities, the outer layer of aluminum was scraped from the cladding of a typical new enriched uranium fuel plate. Approximately 100 milligrams were removed from a surface area of about 15 square inches. The sample was then divided into two portions and irradiated for 17 hours in the Hole 10 facility of the ORNL Graphite Reactor.

After irradiation, the samples were dissolved in hydrochloric acid and chemically processed. The radionuclide 2.33 d Np^{239}, formed by the reaction

$$U^{238}(n,\gamma)U^{239} \xrightarrow[23.5\ m]{\beta^-} Np^{239}$$

served as a measure of the U^{238} content in the irradiated samples. Comparison of the Np^{239} radioactivity in the samples with that of similarly processed uranium standards showed the aluminum scrapings contained approximately 2 ppm U^{238} or about 8×10^{-3} μg U^{238} per square inch of surface area. Similar analysis of new aluminum prior to its use as a cladding showed it to be free of uranium contamination.

The uranium utilized in the fabrication of the fuel elements is greater than 90% enriched in the U^{235} isotope. It is reasonable to assume that the uranium contamination on the fuel plates is similarly enriched. If the ratio of U^{235} to U^{238} is taken as 10 to 1, the U^{235} contamination on the fuel plates would be approximately 8×10^{-2} μg per square inch.

The LITR core is composed of 26 fuel elements each containing 19 fuel plates each of which has about 144 square inches surface area. Assuming uniform U^{235} contamination of 8×10^{-2} μg per square inch, it can be calculated that there are approximately 5.5 mg U^{235} on the surfaces of the fuel plates making up the reactor core.

The LITR has a fuel loading of 3600 grams U^{235} and operates at a power level of 3 megawatts. On this basis, the power level due to the 5.5 mg U^{235} contamination in the core would be approximately 5 watts. The quantity of fission products reaching the water due to such contamination can be calculated as follows.

As a first example, consider the fission product 21 h I^{133}. The fission yield of I^{133} is 6.5% (Blomeke, 1956). At 5 watts power, the number of atoms per second of I^{133} yielded would be:

$$(5 \text{ watts})(3 \times 10^{10} \text{ fissions} \cdot \text{watt}^{-1} \cdot \text{sec}^{-1})(0.065 \text{ atoms} \cdot \text{fission}^{-1})$$
$$= 9.8 \times 10^{9} \text{ atoms} \cdot \text{sec}^{-1}.$$

The coolant flow rate through the LITR core is 20 gallons per second. At this flow rate, assuming all the I^{133} formed due to fission of the U^{235} contamination on the fuel plates reaches the

water, the activity entering the water in each passage through the core would be:

$$\frac{N\lambda}{\text{Volume}} = \frac{(9.8 \times 10^{9}\ \text{atoms}\cdot\text{sec}^{-1})(9.2 \times 10^{-6}\ \text{sec}^{-1})}{(20\ \text{gal}\cdot\text{sec}^{-1})(3785\ \text{ml}\cdot\text{gal}^{-1})}$$

$$= 1.2\ \text{d/s/ml} = 3.2 \times 10^{-5}\ \mu\text{c/ml}.$$

The flow rate through the bypass purification system is 40 gpm. Experimental evidence has shown that the purification system removes essentially all the I^{133} from the water bypassed through it (See Table VIII).

If the cooling system be represented as shown in Figure 83, where:

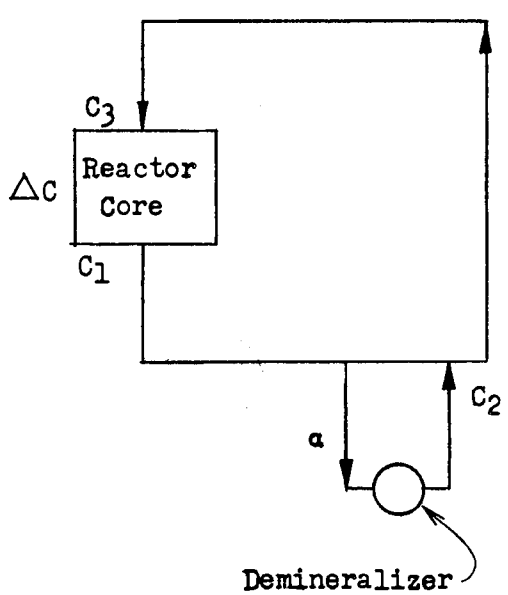

Fig. 83. Simplified Schematic Flowsheet: LITR Cooling Water System.

For the LITR $\begin{cases} t_c = 10\ \text{min} \\ \alpha = 0.033 \end{cases}$

C_1 = concentration of radioactive material in the water leaving the reactor core,

C_2 = concentration of radioactive material in the cooling water just past the point where the purified water from the bypass demineralizer system re-enters the main system,

C_3 = concentration of radioactive material in the water entering the reactor core,

ΔC = gain in concentration of radioactive material in the water during its passage through the reactor core,

α = fraction of total coolant flow bypassed through the demineralizer,

t_c = average time for the water to complete one cycle through the system,

and if the retention time of the cooling water in the reactor core is short compared to the cycle time and if uranium burn up is ignored, then it can be seen (Binford, 1955) that:

$$C_1 = C_3 + \Delta C \qquad (1)$$

$$C_2 = C_1 (1 - \alpha) \qquad (2)$$

$$C_3 = C_2 e^{-\lambda t_c} \qquad (3)$$

$$\text{or} \quad C_3 = C_1(1 - \alpha)e^{-\lambda t_c} \qquad (4)$$

Substituting (4) into (1),

$$C_1 = C_1(1 - \alpha)e^{-\lambda t_c} + \Delta C$$

$$\text{or} \quad C_1 = \frac{\Delta C}{1 - (1 - \alpha)e^{-\lambda t_c}}$$

For small values of α,

$$C_1 \cong \frac{\Delta C}{1 - (e^{-\alpha})(e^{-\lambda t_c})} = \frac{\Delta C}{1 - e^{-(\alpha + \lambda t_c)}} .$$

Where $(\alpha + \lambda t_c)$ is small,

$$C_1 \cong \frac{\Delta C}{\alpha + \lambda t_c}$$

Thus the theoretical concentration of I^{133} in the cooling water due to U^{235} contamination would be:

$$C_1 \cong \frac{\Delta C}{\alpha + \lambda t_c} = \frac{3.2 \times 10^{-5}\ \mu c/ml}{0.033 + (9.2 \times 10^{-6}\ sec^{-1})(10\ min)(60\ sec \cdot min^{-1})}$$

$$= 8 \times 10^{-4}\ \mu c/ml.$$

For comparison purposes, the I^{133} concentration in the influent to the cation exchanger of the bypass purification system can be taken as representative of the concentration in the water leaving the reactor core. The concentration of I^{133} found experimentally in the influent to the cation exchanger was 6×10^{-4} μc/ml (See Table III).

Similar calculations can be made for I^{131} whose fission yield is 2.9% (Blomeke, 1956). The theoretical concentration would be 5×10^{-5} μc/ml. The experimentally found concentration in the influent to the cation exchanger was 8×10^{-5} μc/ml (See Table III).

Thus, the experimental evidence points to the probability that there is adequate U^{235} contamination on or near the surface of the fuel plates to account quantitatively for the fission product concentrations determined in the cooling water. Inasmuch as new aluminum does not contain uranium, it must be assumed that the contamination occurs during fabrication of the fuel elements.

Heath (1956) concluded from studies of the Materials Testing Reactor that the major source of fission products in the cooling water in that reactor was fissionable material distributed on the surfaces of the reactor core structure, such contamination resulting from ruptures of fuel elements and failures of experiments containing fissionable materials. This conclusion need not be in opposition to that reached here. It is quite probable that a fuel element rupture in a high power reactor of the MTR type so contaminates the reactor vessel that uranium contamination possibly present on or near the

surface of new fuel elements becomes insignificant.

C. Induced Radioactive Nuclides

1. Na^{24}

Of the radioactive nuclides thought to originate from induced activity, Na^{24} is of the most importance. Relatively large quantities have been observed in the LITR cooling water since initial operation (Rupp and Cox, 1956). Early studies as to the source of the Na^{24} were conducted by Binford and Lyon. Through measurement of the variation in the 15.0 h Na^{24} content in the cooling water for various bypass purification rates, they sought to determine whether or not the radioactive sodium originated from neutron irradiated stable elements in the water. Their data, although not conclusive, strongly indicated that the majority of the Na^{24} came from sources other than stable elements in the water (Binford, F. T. and Lyon, W. S., 1957).

Heath (1956), through studies of the Na^{24} content of the cooling water of the MTR at different power levels, concluded that the major source of radioactive sodium was the $Al^{27}(n,\alpha)Na^{24}$ reaction. Heath also found a much lower Na^{24} content in cooling water circulating in stainless steel and carbon steel systems than in those employing aluminum clad fuel.

Quantitative neutron activation analysis (Leddicotte and Reynolds, 1954) of the demineralized make-up water for the LITR cooling system showed it to contain 2.3×10^{-2} ppm stable sodium. A similar specific analysis of the effluent cooling water from the

mixed-bed exchanger showed the content of stable sodium at this stage to be 6.0×10^{-3} ppm. Using the average thermal neutron flux and retention time of the water in the core, an estimate can be made of the amount of 15.0 h Na^{24} which would result from these amounts of stable sodium in the circulating coolant. Calculations assuming a total daily retention time of 108 seconds and an average thermal flux of 2.2×10^{13} neutrons$\cdot cm^{-2} \cdot sec^{-1}$ show that these amounts of stable sodium would account for only approximately one tenth to one hundredth of the Na^{24} routinely found in the water.

Thus, these data would appear to further substantiate aluminum as the principal source of Na^{24} in the cooling water. This includes aluminum in the fuel cladding as well as that dissolved in the water. The air coolers and part of the piping are aluminum, there being a total of about 3,000 square feet of exposed aluminum in the cooling system. Spectrographic analysis of the make-up water and the mixed-bed effluent showed both to contain approximately 0.02 ppm aluminum.

b. $\underline{Cu^{64}}$

The second important nuclide in the cooling water is 12.8 h Cu^{64}. The quantity of this nuclide increased considerably near the conclusion of these studies. Since its most probable source was stable copper, a check was made of the copper components in the cooling system. It was known, for example, that the mounts for the clarifying filters on the influent and effluent sides of the bypass purification system were of copper construction. These units, upon inspection, showed pronounced evidence of corrosion.

Replacement of the copper mounts with similar units of stainless steel caused an immediate reduction in the amount of Cu^{64} in the water.

The effect of such corrosion was also reflected in the ion exchange resins. Whereas the cation bed resin sample of December 28, 1956, showed Na^{24} to be the principal radioactive nuclide retained on the upper portion of the bed, the predominant activity on a similar sample taken just before replacement of the filter mounts was Cu^{64}. This incident as well as the experience concerning the passage of I^{131} through the anion bed emphasizes the value of routine sampling of cooling water purification systems. Such systems, as these data show, are dynamic, being influenced by many variables and constantly changing as these influences have their effect.

3. F^{18}

The source of this radionuclide was discussed in Chapter VI.

4. Mo^{99}, Co^{60}, Mn^{56}, Sb^{122}, Cr^{51} and Fe^{59}

Most all the remaining radioactive nuclides identified in the water probably originate from neutron activation of stable elements. One source of the stable elements is the make-up water, itself. A second source is structural material within the reactor system. For example, the water in the cooling system is exposed to large amounts of steel surfaces. These are in the reactor tanks, valves, pump housing, fittings and a portion of the pipe. In addition, the demineralizer system, the pump shafts and the impellers

are of stainless steel, the total area of exposed steel in the system being about 700 square feet (Rupp and Cox, 1956). Common impurities in steel include molybdenum, manganese and chromium.

Some radionuclides induced in structural materials probably gain access to the water through recoil at the instant of formation. Others may originate through corrosion whereby either the radioactive nuclide gains access to the water or the stable nuclide gains access only to be activated through neutron irradiation at a later time.

D. Summary

Table XII contains a list of all the radionuclides identified in the LITR cooling system with their probable modes of formation.

TABLE XII

Probable Modes of Formation of Radioactive Nuclides Identified in LITR Cooling Water System

Radionuclide	Primary Mode of Formation	Secondary Mode of Formation
Ba^{140}	Fission Product	
Br^{80}	$Br^{79}(n,\gamma)Br^{80}$	$Br^{80m} \xrightarrow{IT,\gamma} Br^{80}$
Br^{80m}	$Br^{79}(n,\gamma)Br^{80m}$	
Br^{82}	$Br^{81}(n,\gamma)Br^{82}$	
Ce^{141}	Fission Product	$Ce^{140}(n,\gamma)Ce^{141}$
Ce^{143}	Fission Product	$Ce^{142}(n,\gamma)Ce^{143}$
Co^{60}	$Co^{59}(n,\gamma)Co^{60}$	$Cu^{63}(n,\alpha)Co^{60}$
Cr^{51}	$Cr^{50}(n,\gamma)Cr^{51}$	
Cu^{64}	$Cu^{63}(n,\gamma)Cu^{64}$	$Zn^{64}(n,p)Cu^{64}$
F^{18}	$O^{18}(p,n)F^{18}$	
Fe^{59}	$Fe^{58}(n,\gamma)Fe^{59}$	$Co^{59}(n,p)Fe^{59}$
I^{131}	Fission Product	
I^{132}	Fission Product	
I^{133}	Fission Product	
I^{135}	Fission Product	
La^{140}	Fission Product	$La^{139}(n,\gamma)La^{140}$
Mn^{56}	$Mn^{55}(n,\gamma)Mn^{56}$	$Fe^{56}(n,p)Mn^{56}$
Mo^{99}	$Mo^{98}(n,\gamma)Mo^{99}$	Fission Product
Na^{24}	$Al^{27}(n,\alpha)Na^{24}$	$Na^{23}(n,\gamma)Na^{24}$
		$Mg^{24}(n,p)Na^{24}$

TABLE XII (continued)

Probable Modes of Formation of Radioactive Nuclides Identified in LITR Cooling Water System

Radionuclide	Primary Mode of Formation	Secondary Mode of Formation
Np^{239}	$U^{238}(n,\gamma)U^{239}$	
	$U^{239} \xrightarrow{\beta^-} Np^{239}$	
Pa^{233}	$Th^{232}(n,\gamma)Th^{233}$	$U^{235}(n,\gamma)U^{236}$
	$Th^{233} \xrightarrow{\beta^-} Pa^{233}$	$U^{236}(n,\gamma)U^{237}$
		$U^{237} \xrightarrow{\beta^-} Np^{237}$
		$Np^{237} \xrightarrow{\alpha} Pa^{233}$
Sb^{122}	$Sb^{121}(n,\gamma)Sb^{122}$	
Sr^{91}	Fission Product	
Sr^{92}	Fission Product	
Tc^{99m}	$Mo^{98}(n,\gamma)Mo^{99}$	Fission Product
	$Mo^{99} \xrightarrow{\beta^-} Tc^{99m}$	
Te^{132}	Fission Product	
Xe^{131m_1}	Fission Product	
$Xe^{133} + Cs^{133m}$	Fission Product	$Xe^{132}(n,\gamma)Xe^{133}$
$Xe^{135} + Cs^{135m}$	Fission Product	$Xe^{134}(n,\gamma)Xe^{135}$
Y^{91m}	Fission Product	
Zn^{65}	$Zn^{64}(n,\gamma)Zn^{65}$	$Cu^{65}(p,n)Zn^{65}$

CHAPTER X

SUMMARY AND CONCLUSIONS

Cooling systems are dynamic, being influenced by many variables and constantly changing as these influences have their effect. Data regarding the nature and levels of radionuclides in the cooling water readily reflect unusual conditions in the reactor system. Such conditions may include fuel element rupture, failure of experimental facilities and excessive corrosion. As a result, routine monitoring of the coolant is a necessity for safe, as well as efficient, reactor operation.

Studies of the cooling water system of the Low Intensity Test Reactor at the Oak Ridge National Laboratory substantiated these observations. Radionuclides identified in the cooling system of this reactor included Na^{24}, Np^{239}, Cu^{64}, I^{131}, I^{132}, I^{133}, I^{135}, $Mo^{99} + Tc^{99m}$, Mn^{56}, $Sr^{91} + Y^{91m}$, F^{18}, Sb^{122}, Pa^{233}, $Ba^{140} + La^{140}$, Co^{60}, Ce^{143}, Ce^{141} and Te^{132}. These were of two groups: (a) fission products and (b) induced activities.

It has been shown that there was adequate uranium contamination on or near the surface of the aluminum cladding of typical new fuel plates to account quantitatively for the fission products in the coolant. Such contamination probably occurs during fabrication of the elements. Secondary sources of fission products may arise through pitting of fuel plates and spills of experimental uranium samples within the reactor vessel.

The induced activities were thought to originate through neutron activation of reactor structural materials and of impurities in the make-up water. Various nuclides in the structural materials could gain access to the water, either before or after activation, through corrosion processes. For example, unusual Cu^{64} contamination in the cooling water was found traceable to corrosion of copper filter mounts. Replacement of the mounts with those of a stainless steel type caused an immediate reduction in the contamination in the system. This approach to control of radionuclide concentrations in the cooling water represents the ideal, that is, control at the source. It is probable that similar source control of fission product contamination can be accomplished through use of non-contaminated fuel elements.

For certain radionuclides, control at the source is impractical or impossible. For these, the ion exchange purification system must act as control. It has been shown that the ion exchange system employed in the LITR is effective in removing radionuclides from the cooling water. Maintenance of high purity water in the reactor system has had many advantages. These include lowered radiation intensities surrounding the cooling water pumps and piping, reduced contamination on tools used within the reactor tank and reduced accessibility problems. In addition, the use of water of high specific resistance has resulted in much lower corrosion rates -- an important factor in the life of the reactor (Rupp and Cox, 1956).

The effectiveness of the control methods in the LITR cooling system eliminates the need for the release of radioactive material to the environment through cooling water discharge. Radioactive material removed from the water is retained on the ion exchange resins until removed by regeneration. The small volume of regenerating solutions is adaptable to storage. Storage of the large volumes of water discharged from reactors employing the single pass system is impractical. Consequently, in the case of fuel element rupture or other accidents releasing radionuclides into the coolant stream, there is the possibility that significant amounts of radioactive material could be discharged into the environment. In general, therefore, from the public health point of view, the recirculating system is the type to be recommended.

In the studies reported herein, certain radiochemical and radioactivity assay techniques were modified for specific application to water analysis. These techniques have significance beyond the field of reactor cooling water. For example, neutron activation analysis could find routine application in the identification and determination of stable elements in drinking water. In many studies of drinking water impurities today, health agencies are interested in measuring extremely small concentrations. For certain elements, the sensitivity of the activation analysis technique is unparalleled.

The successful use of the gamma scintillation spectrometer in the determination of radionuclides in water in quantities near current maximum permissible limits has been demonstrated. This instrument

could find application in the identification and determination of radionuclides in streams and lakes, the contamination of which is of current interest to many state health departments.

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