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A FIELD INSTRUMENT FOR QUANTITATIVE DETERMINATION OF BERYLLIUM BY ACTIVATION ANALYSIS

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FIELD INSTRUMENT FOR QUANTITATIVE DETERMINATION OF BERYLLIUM BY ACTIVATION ANALYSIS

By W. W. Vaughn, E. E. Wilson, and J. M. Ohm

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ABSTRACT

A low-cost instrument has been developed for quantitative determinations of beryllium in the field by by activation analysis. The instrument makes use of the gamma-neutron reaction between gammas emitted by an artificially radioactive source (Sb^{124}) and beryllium as it occurs in nature. The instrument and power source are mounted in a panel-type vehicle. Samples are prepared by hand-crushing the rock to approximately $\frac{1}{4}$-inch mesh size and smaller. Sample volumes are kept constant by means of a standard measuring cup. Instrument calibration, made by using standards of known BeO content, indicates the analyses are reproducible and accurate to within ± 0.25 percent BeO in the range from 1 to 20 percent BeO with a sample counting time of 5 minutes. Sensitivity of the instrument may be increased somewhat by increasing the source size, the sample size, or by enlarging the cross-sectional area of the neutron-sensitive phosphor normal to the neutron flux.

INTRODUCTION

In February of 1958, the U.S. Geological Survey began experimental work with an activation analysis technique using photoneutrons to indicate the presence of beryllium in rock samples. The object of the work was to develop a low-cost reliable beryllium assay instrument that would require minimum sample preparation and analytical time. During the course of the development of the described instrument, the authors became aware of a similar development program of an instrument for field exploring by Dr. George M. Brownell of the Geology department of the University of Manitoba, Winnipeg, Canada, through a visit by Dr. Brownell to the Geological Survey offices in Denver, Colorado. A description of Dr. Brownell's instrument has been published (Brownell, 1959).

When a gamma source emitting photons with an energy of at least 1.66 Mev is placed in close proximity to a material containing beryllium, neutrons are released from the nuclei of the beryllium atoms. This reaction, commonly expressed as $Be^9(\gamma, n)Be^8$, has been used for at least two decades in applications where radium and beryllium in concentrated form are combined to provide a source of neutrons for experimental work with nuclear-induced reactions. Determinations of the $Be^9(\gamma, n)Be^8$, reaction using γ-rays from the radium chain and from ThC"(Tl^{208}) were summarized by Houtermans and Bartz in 1943. In 1948, Russel, and others, described experiments using a small chamber whose walls were composed principally of beryllium. The chamber was filled with various gamma emitters and the neutron flux from the beryllium was measured. Russell noted the high efficiency with which neutrons were produced when antimony and beryllium were placed in close proximity. He concluded that there was probably a sharp peak in the yield curve at the photon energy of antimony, because at higher photon energy levels the yield declined. Results of this experimental work indicated that under favorable conditions, and with a good neutron detector, the presence of beryllium in nature could be measured if the geometric relation between the irradiating photon flux and the beryllium-bearing material were optimum.

In 1950, Gaudin and others reported on the use of this nuclear reaction in the design of an instrument for automatic concentration of beryl. A one-curie radium source, emitting approximately 30 percent of its photons with energies above 1.66 Mev, (the threshold level for the production of neutrons in beryllium), was used as the irradiating source. A boron trifluoride (BF_3) gas-filled counter tube was used to detect the neutrons from the beryllium after they are thermalized by paraffin. With this arrangement, a counting-rate ratio between the background from the radium gamma source and the neutrons originating in beryllium of 1:5 or better was possible. Considerable shielding and proper handling precautions are necessary for personnel safety when using a one-curie source of radium. The high penetration character of gamma radiation minimizes the problem of uniform preparation and positioning of sample material, which is desirable when quantitative results are required.

A nuclear detector for beryllium described by Cantwell and others (1958) used the nuclear reaction between polonium alphas and the nucleus of a beryllium atom.

$$_4Be^9 + {}_2He^4 \longrightarrow {}_6C^{12} + {}_0N^1$$

(Alpha; neutron)

The alpha-neutron (α, n) reaction was discovered by Chadwick in 1932. Combined concentrations of Po and Be are also used for commercial neutron sources ranging up to several curies in strength. The capture cross section for the α, n reaction is approximately 250 millibarns as compared to 1 millibarn for the gamma-neutron (γ, n) reaction. The barn (10^{-24} sq cm) in a "cross sectional" measurement of the nucleus of an atom and is a measure of its probability of absorbing incident radiation in terms of the type and energy of the incident radiation. The threshold level for the release of the neutron from a beryllium atom by an alpha particle is 3.7 Mev.

Because the probability of a neutron being released in beryllium with the α, n reaction is 250 times greater than in the γ, n reaction, and selected alpha sources have very little interfering gamma activity, the α, n reaction would seem more desirable for beryllium determinations. However, with the α, n technique, the alpha emitter (usually polonium) must be plated and

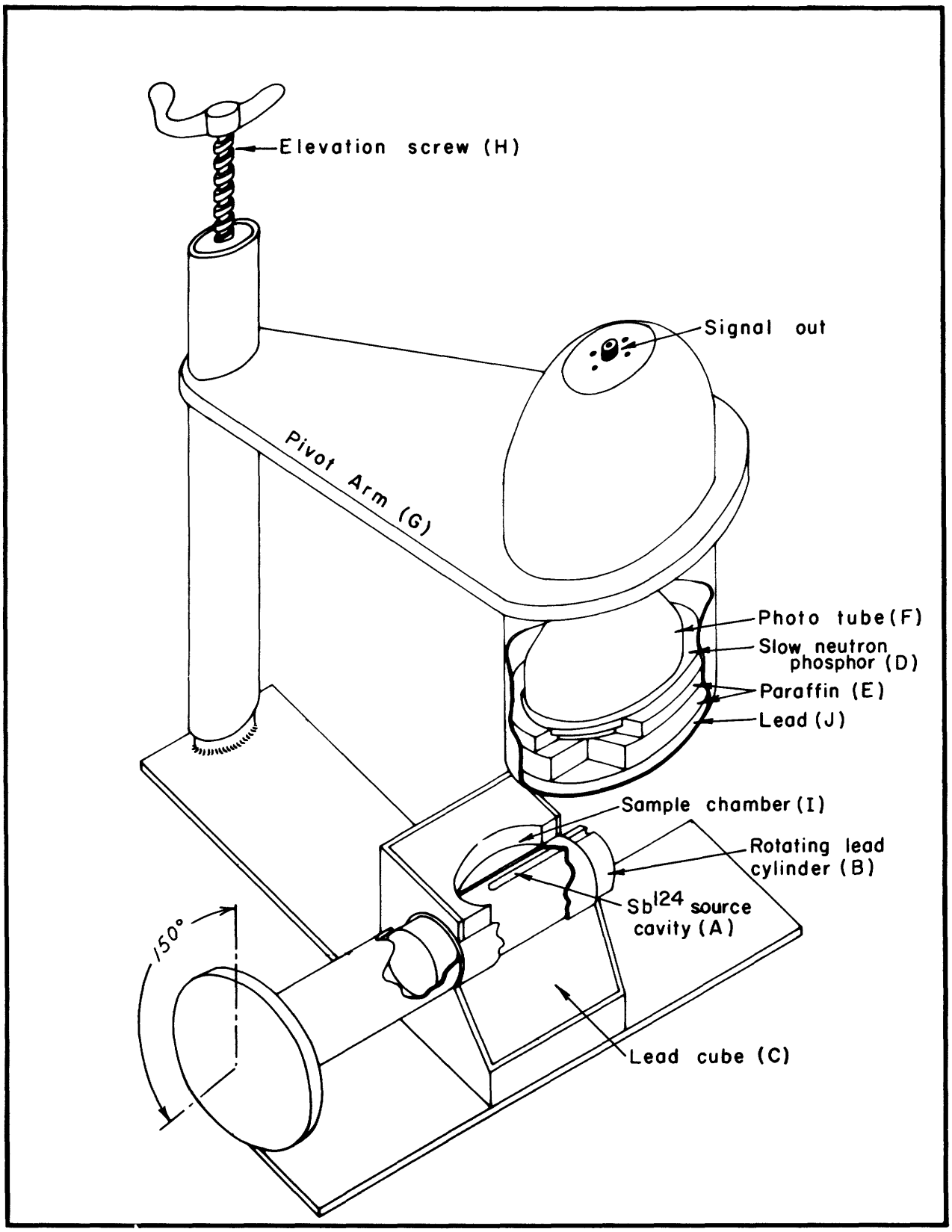

Figure 1.—Beryllium detector for quantitative determinations.

sealed on a surface with a window 1 mill or less in thickness and essentially transparent to the alpha radiation. With the alpha source very close to this window, only a thin layer on the surface of a finely powdered sample can be analyzed. Because the window containing the polonium is by necessity very fragile, frequent monitoring with an alpha-sensitive instrument is required to insure against leakage of the sealed polonium source.

The γ,n reaction was selected primarily because it affords greater flexibility for nonuniform sample preparation in field work. Personnel safety is assured through simple dosimetry procedures.

ACKNOWLEDGEMENTS

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PHYSICAL DESIGN

The instrument was designed mechanically to provide a simple and rapid means of analyzing beryllium-bearing material in the field (fig. 1) The Sb^{124} source is inserted in an opening (A) on the perimeter of, and parallel to, the axis of a lead cylinder (B) approximately 8 inches long and 3 inches in diameter. The lead cylinder is then placed in a brass-lined opening in a 6-inch lead cube (C). This cylinder can be turned through an arc of 150°, allowing the gamma-emitting source to be rotated from the shielded position in the lead container to the irradiation position for sample analysis with a minimum of exposure to personnel. The scintillation head containing the boron-zinc sulfide thermal neutron detector (D), the paraffin moderator (E) for thermalizing the neutrons from the beryllium, the photomultiplier tube (F), and associated circuitry are mounted on a pivot arm (G) above the sample chamber. A hand-operated jackscrew (H) provides a means of adjusting the height of the sensitive head above the sample chamber (I). With this provision a large, irregularly shaped rock can be examined qualitatively for the presence of beryllium without crushing the specimen.

When the Sb^{124} source is rotated to the exposed position for counting two lobe-shaped cams, which are an integral part of the shaft for the lead cylinder containing the source, are aligned on either side of the sensitive head and extend above the plane of travel of the bottom of the chamber. The sensitive head with the inch-thick lead plate (J) in the lower compartment is therefore locked in position while the analysis is being run. This precaution prevents the sensitive head and the protective lead plate from being swung aside, exposing the operator to the full flux from the Sb^{124} gamma source. A rocker arm operated by the lateral movement of the sensitive head locks the source holder in the "safe" position when the chamber is swung aside for insertion of the sample in the sample chamber. The assembly is fastened securely to a $\frac{1}{4}$-inch steel base plate which is attached to the floor of the vehicle.

ELECTRICAL DESIGN

In the activation-analysis technique, it is necessary that the gamma-activating source be close to the sample material and, except in rare applications, relatively close to the neutron detector.

All neutron detectors are somewhat gamma sensitive. Therefore, it is desirable to have a detector with a large neutron to gamma sensitivity ratio. A boron-enriched (13 percent B^{10} by weight) polyester containing zinc sulfide was selected. This phosphor, in addition to being a good neutron detector, permits discrimination against gammas by pulse height selection which is an inherent advantage in all scintillation phosphors.

If a linear, light-amplifying device such as the photomultiplier tube is used to observe the scintillations occuring in the phosphor, the voltage pulse developed at the anode of the photomultiplier tube will be proportional to the intensity of the light flash in the phosphor. The light flash, within reasonable limits, is proportional to the type and amount of energy dissipated in the phosphor by the incident radiation.

A minimum amount of circuitry is used in the sensitive element. The photomultiplier tube (fig. 2) is operated with the cathode grounded and the anode at a high positive potential. The voltages for dynodes 1 through 10 are supplied from a voltage divider consisting of a series of one megohm resistors. Dynode 10 is by-passed to the cathode by a 0.01 microfarad 1,600-volt capacitor to maintain a constant voltage across the dynode string. The amplification factor of the photomultiplier is determined by the voltage applied, and is set at a value that gives the desired response. The overall sensitivity of the instrument is therefore adjusted by varying the voltage across the dynode string.

The transparent side of the plastic neutron phosphor is mounted directly on the cathode face of the photomultiplier tube to give maximum optical coupling. A thin film of clear silicone grease is applied to the face of the photomultiplier before the the phosphor is brought into contact with the glass. The two surfaces are then joined and revolved slightly to force out captured air bubbles.

The pulse spectrum appearing at the anode of the photomultiplier tube as a result of the light flashes in the phosphor is shown in figures 3 and 4. Figure 3 is a photograph of gamma activity with the Sb^{124} source in the counting position and the beryllium sample removed from the instrument. Figure 4 is a photograph of the gamma and the neutron flux with a sample of beryl in the counting chamber. The oscilloscope gain-control setting and the film-exposure time were the same for both photographs. Neutron activity results in many pulses of light of approximately the same amplitude as those caused by the gamma radiation. There is, however, a sufficient quantity of high-amplitude neutron pulses (figs. 5 and 6) to permit the input bias to be set at a level at which no gamma pulses will be accepted by the counting circuit. Therefore, the use of pulse-height-selection techniques makes possible an accurate recording and correlation of counting rate, as a function of beryllium content of the sample. The only interference in this pattern is due to cosmic neutrons (the largest pulse in figure 4) which, for the described instrument, amounts to approximately six counts per

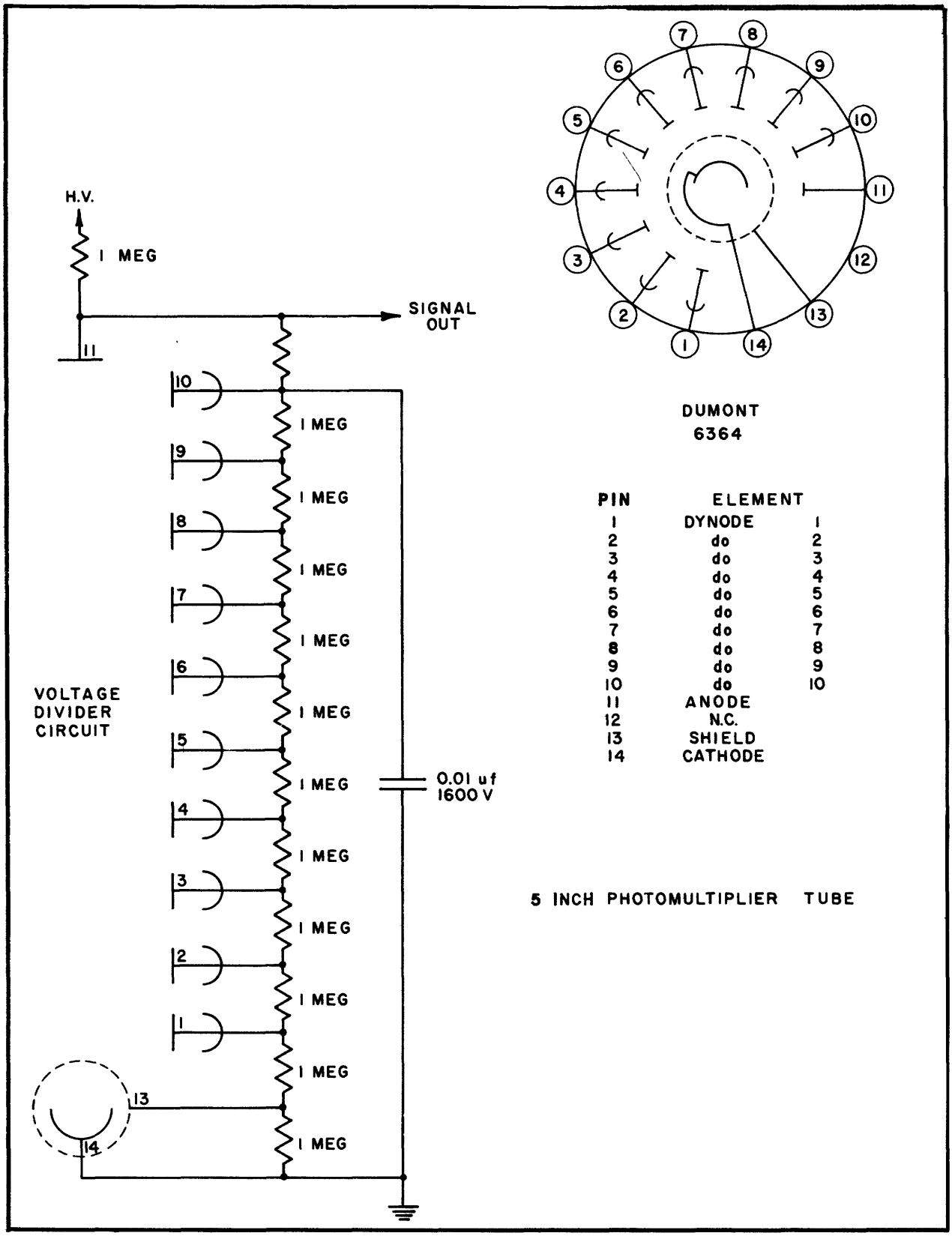

Figure 2.—Photomultiplier tube circuitry.

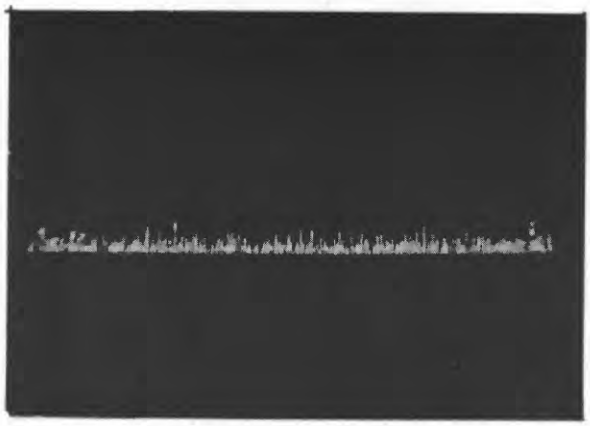

Figure 3.—Oscilloscope trace of gamma background from Sb^{124} with beryllium sample removed.

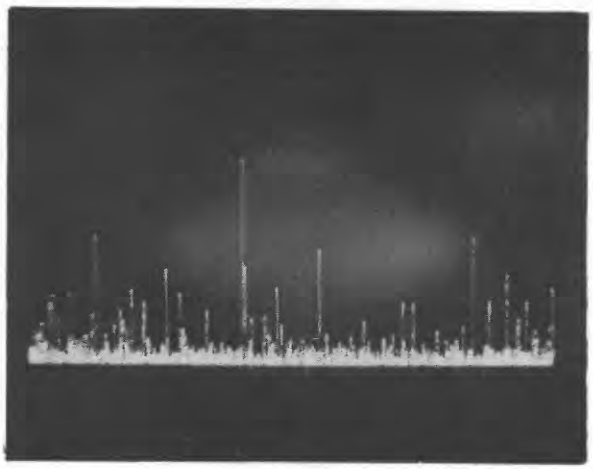

Figure 4.—Oscilloscope trace of gamma background from Sb^{124} and neutron flux with flux beryllium sample in place.

minute at 6,000 feet above sea level. It is possible to eliminate this cosmic background with additional pulse height analysis, however, it was preferred to subtract the cosmic events from the gross counting rate because the number is small and seems to remain rather constant over a period of time.

The photomultiplier tube and its operational characteristics are critical when used with this technique. For best instrument performance, the operational characteristics of the particular photomultiplier tube used should be known. A graph (fig. 5) of counting rate as a function of voltage can be plotted without a

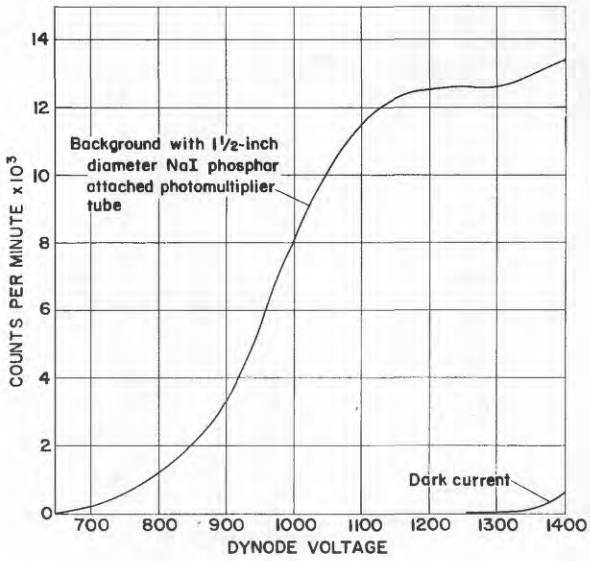

Figure 5.—Operational characteristics of a photomultiplier tube.

sample in the counting chamber. This graph shows the high voltage value at which high voltage drift has a minimum effect on counting rate. The phosphor, or any other source of light that might activate the photomultiplier, can then be removed, the high voltage settings repeated, yielding a plot of the dark-current noise factor, (fig. 5). A logical operating high voltage on the plateau and below the dark current region would be 1,225 volts for the curves shown.

The average light emitted by the plastic neutron phosphor can be raised to such a high level by excessive gamma radiation, that additional pulses from the neutron flux have no effect, and the instrument becomes inoperative. In practice, it has been found that the gamma field from the Sb^{124} source at the phosphor should not exceed 1,000 milliroentgens per hour. The phosphor may also be temporarily "saturated" by normal room light. When the entire phosphor is exposed to room light for several minutes, the phosphor will be energized to a light level that is easily observed with the naked eye in a dark room.

Using the circuitry shown in figure 2, the amplitude of the pulses resulting from neutrons interacting with the phosphor range from 10 to 250 millivolts, the largest number occurring between 20 and 50 millivolts. Pulse height of 20 to 50 millivolts are adequate to trigger most commercially available scalers or rate meters that might be used as counting devices.

INSTRUMENT OPERATION

An operational schematic of the beryllium detector is shown in figure 6. The gamma emitter (100 millicuries of Sb^{124}) has a half life of 60 days. Approximately 70 percent of the photons emitted have an energy of 1.7 Mev which is sufficiently above the 1.66 Mev threshold energy level for the production of neutrons in beryllium.

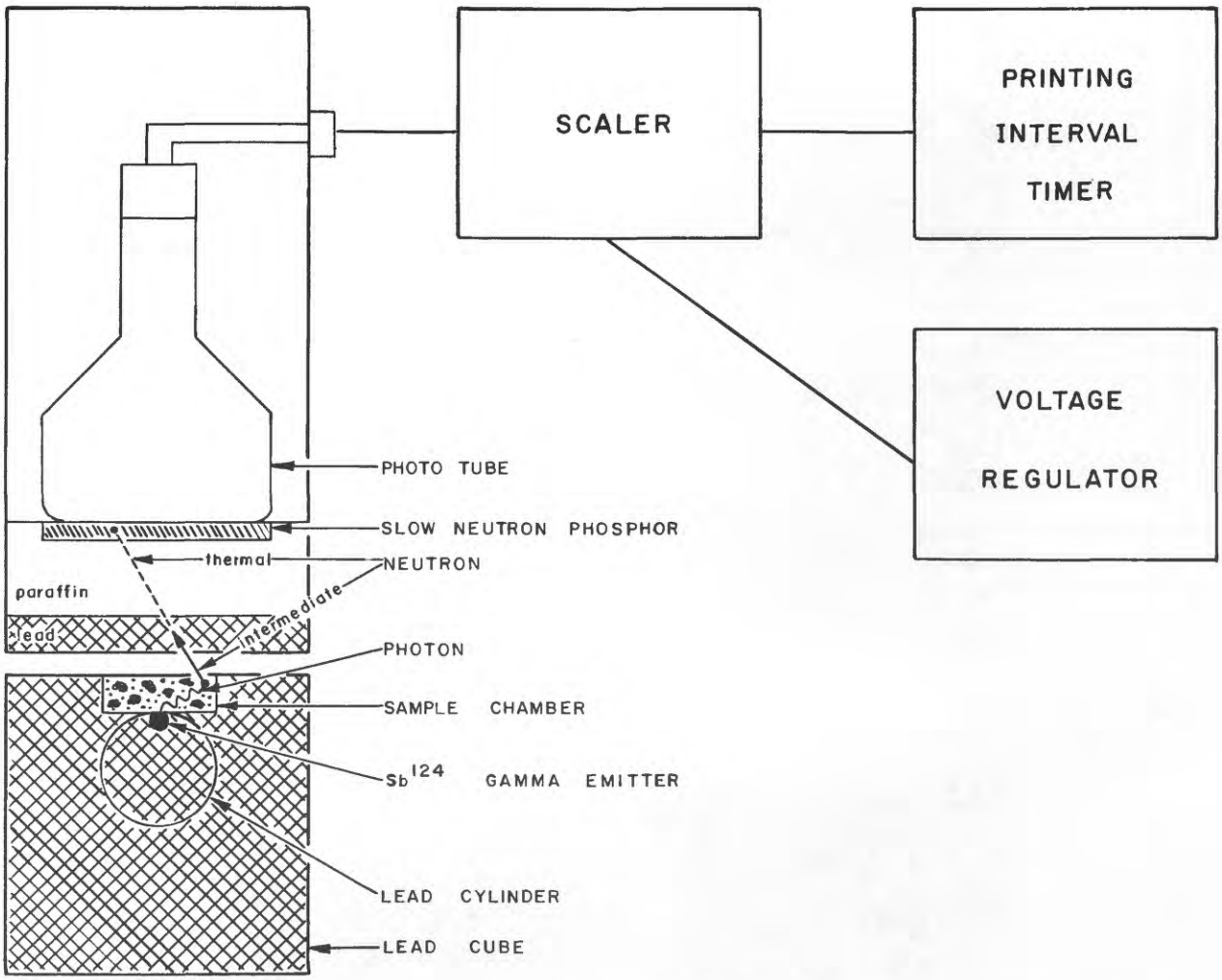

Figure 6.—An operational schematic of the beryllium detector.

The neutrons originating in the beryllium have an energy centered around 0. 24 Mev region, and and are considered to be in the intermediate energy range. Although the neutrons originating in the beryllium cannot be considered as truly monoenergetic, the energy spread is probably less than 20 Kev. This makes it possible to slow down, or thermalize most of the neutrons released from beryllium.

The neutron has great penetrating power because it has no charge and therefore does not ionize atoms by dislodging electrons from their orbits as charged particles do. It will lose energy to atomic nuclei by a series of elastic collisions.

Once the progress of the neutron has been slowed or moderated to the thermal excitation level of the medium traversed, it may either lose or gain energy with successive collisions with nuclei until it is captured by the nucleus of an atom.

In the equipment, described paraffin is used as the moderator from which the neutrons emerge with about 0.025 ev energy and are, on the average, captured by the nucleus of B^{10} atom on entering the slow neutron phosphor. On absorbing the neutron, the B^{10} atom emits an alpha particle which reacts with the ZnS in the phosphor and creates a flash of light. This scintillation, in turn, excites the photosensitive cathode surface of the photomultiplier tube which releases a cloud of electrons. Through secondary emission, from dynode to dynode, the cloud of electrons is amplified until a sufficient current density is realized at the anode to form a voltage pulse across the 1 megohm anode load resistor.

SAMPLE PREPARATION

The following method has been used and, although seemingly crude, has provided consistent results within

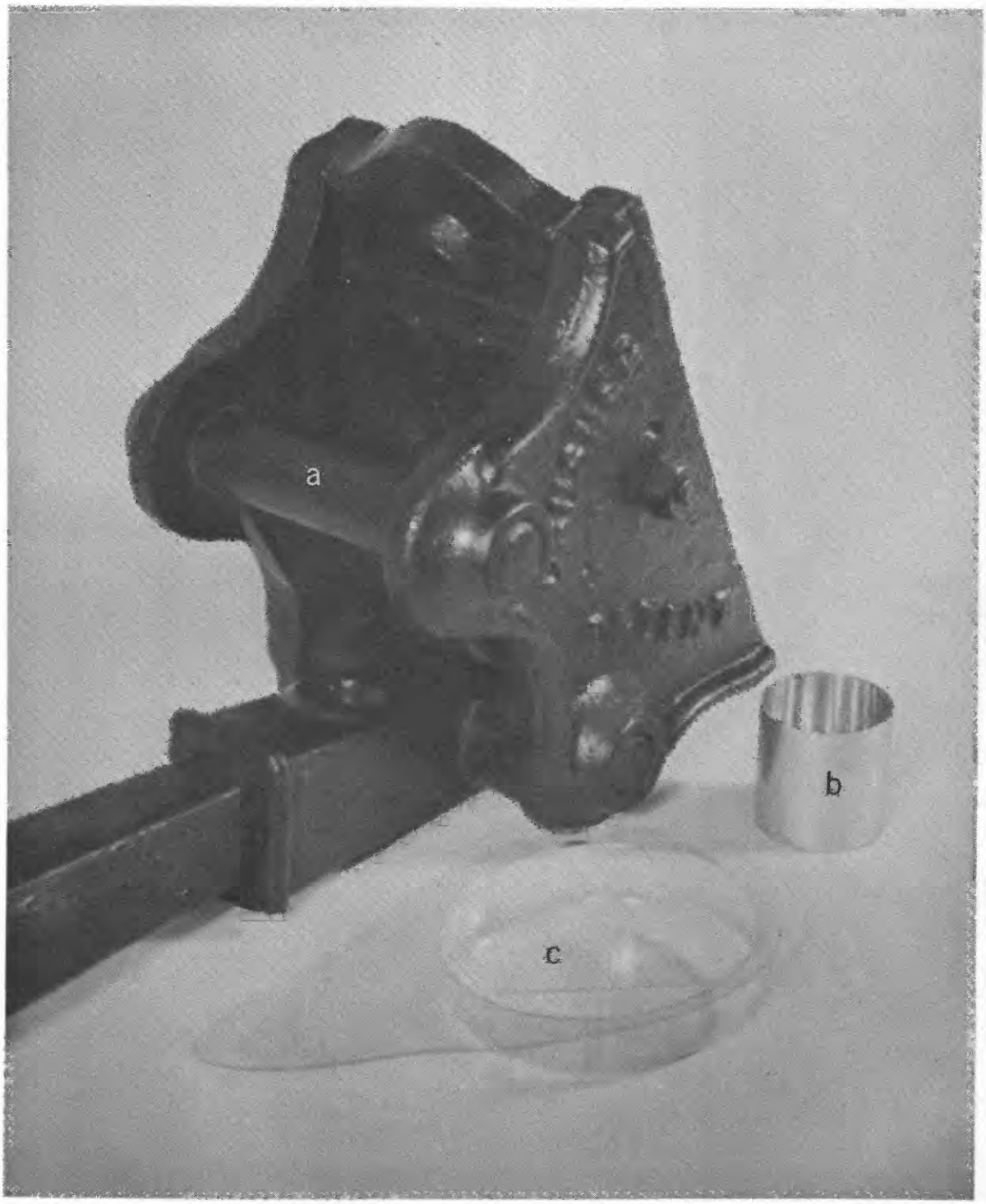

Figure 7.—Equipment for sample preparation in the field for quantitative beryllium determinations.

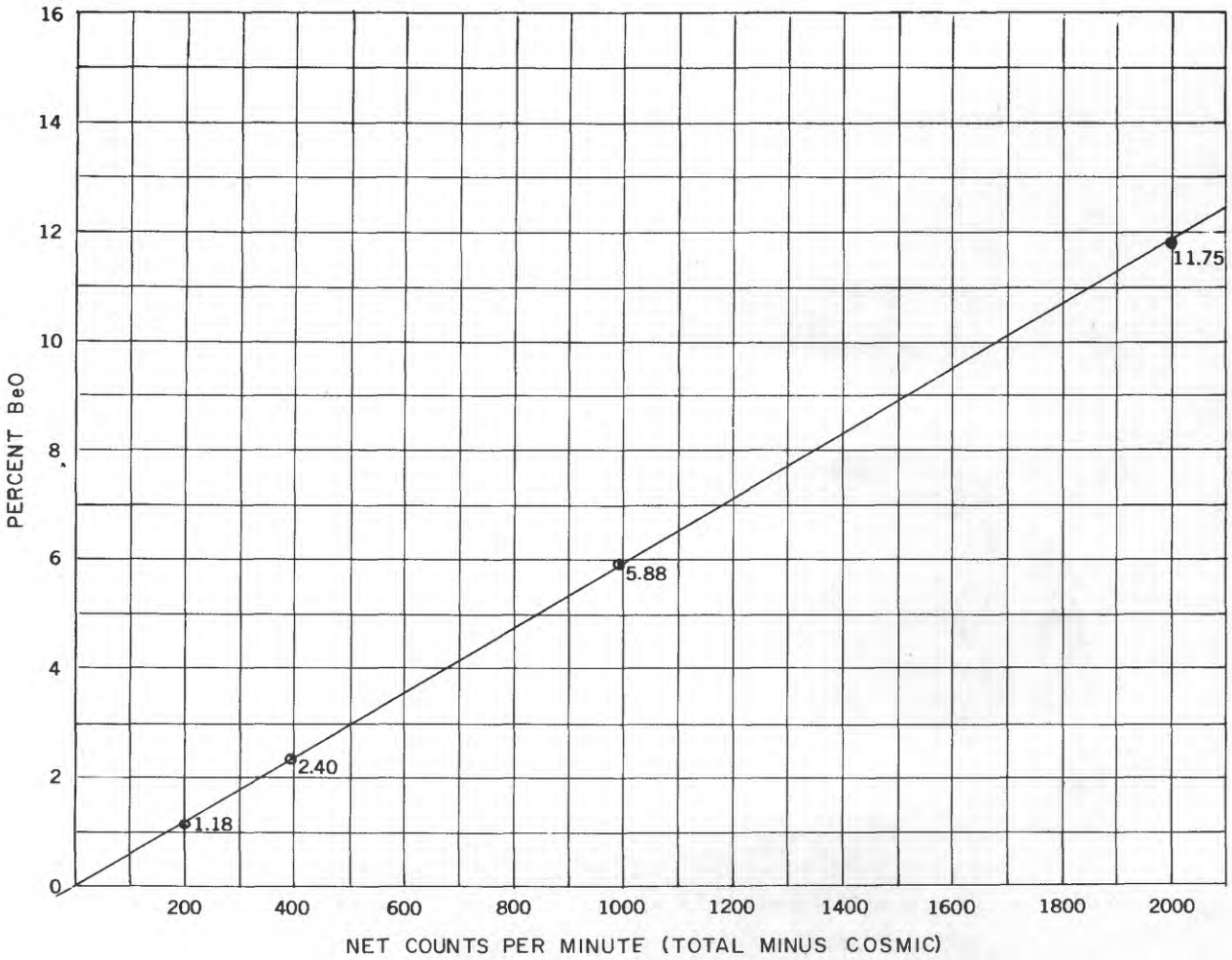

Figure 8.—Calibration curve for the beryllium detector.

the operation error (approximately ± 0.25 percent BeO) of the equipment under field conditions. For analysis below 1 percent BeO, a more refined system should be devised.

Figure 7 shows the equipment for sample preparation in the field. The sample is crushed by a commercially available hand-operated crusher (a) which has adjustable jaws for varying the particle size. A split of the crushed material is measured with a standard container (b) which hold approximately 70 grams, and placed in the plastic sample holder (c). The plastic sample holder, made by the vacuum-mold method, is 0.020-inch-thick acetate sheet, 3 inches in diameter and $\frac{3}{4}$-inch deep.

To make the sample holder, a 6-inch square of the acetate sheet is held between two frames of aluminum hinged in book form and is heated uniformly to 100°C. The assembly is then removed from the furnace and immediately placed over a machined mold from which air is drawn through a series of small holes. The plastic material, while it is still pliable, is quickly drawn down and conforms exactly to the mold shape. By inverting the mold, a second container can be made which serves as the top of the first molding. An ordinary vacuum cleaner provides a sufficient vacuum for this purpose.

CALIBRATION

Figure 8 shows a typical calibration curve for the beryllium detector. Standards for this calibration were prepared by mixing weighed amounts of ground quartz and beryl; the beryle was taken from a crystal that had been chemically analyzed. An average net counting rate (total minus cosmic) was obtained for each sample and plotted as a function of the BeO content. The counting rate increases linearly with the amount of beryllium in the sample. In the range 1 to 20 percent BeO, a counting time of 5 minutes was sufficient to give satisfactory reproducibility for field samples.

Rock samples may have concentrations of boron, lithium, and other elements with a relatively high absorption cross section for neutrons. It was realized that presence of these elements might seriously affect the instrument readings. However, tests run with prepared samples containing 17 percent of boron

by weight gave no appreciable deviation in counting rate from a boron-free split of the same sample material. This probably results from the small sample size and from the low specific energy loss of the intermediate range neutrons in the sample material.

To study the effects of grain size, a count was taken on a measured amount of various size fractions of a given sample, ranging from $\frac{1}{4}$-inch and all fines down to a rather consistent grind passing 100 mesh. There was no significant variation in data in the range of sensitivity studied due to grain size.

The half-life of Sb^{124} is 60 days. Therefore, assuming 100 percent initial activity, after 60 days 50 percent of the activity will remain and after another 60 days, half of the 60-day value or 25 percent of the original activity, and so on as time progresses. This decay must be taken into account in the quantitative calibration of beryllium detectors over the period of time the instrument is used. As the activity decreases, the slope of the calibration curve (as plotted in figure 8) will increase, that is, the number of neutrons produced by a given beryllium concentration decreases. Probably the simplest way to correct for the half-life decay is to establish a new calibration curve each week, or more often as circumstances require. This procedure not only establishes a calibration directly associated with the instrument and its environment, but provides a permanent record of overall instrument operational effectiveness. A 100-millicurie source of Sb^{124} has given satisfactory analyses in the range 0.5 to 12 percent. BeO over a period of approximately 10 months (5 half lives).

POSSIBLE APPLICATION

The gamma-neutron reaction for beryllium analysis using scintillation counting techniques may be adapted to prospecting, core scanning, drill-hole logging, and to other applications where radiometric methods apply. The most extensive use of the technique, however, will probably be made in mines and in process control. Under optimum conditions and with a large activating source, the instrument could provide instantaneous and continous monitoring of the beryllium content of a slurry or fine aggregate. The beryllium content could be indicated automatically in the form of voltage or current. The monitoring signal could in turn be used to actuate an electromechanical device for the automatic control of the process. Another possible application would be in conjunction with air pollution in an industrial environment where beryllium is being processed. By passing the air through the appropriate filter paper and then passing the filter paper through the counting chamber, an indication of the beryllium contained in the air could be obtained.

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