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LOS ALAMOS SCIENTIFIC LABORATORY
OF THE UNIVERSITY OF CALIFORNIA o LOS ALAMOS NEW MEXICO

QUANTITATIVE MEASUREMENT OF GAMMA-RAY-EMITTING RADIONUCLIDES IN METEORITES

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QUANTITATIVE MEASUREMENT OF GAMMA-RAY-EMITTING RADIONUCLIDES IN METEORITES

by

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ABSTRACT

The gamma-ray spectra of 32 samples of 25 chondrites, 4 carbonaceous chondrites, and 9 achondrites have been investigated using a large NaI (Tl) scintillation spectrometer. Potassium and Al^{26} were the predominant radioactivities measured. More unusual gamma-ray emitters measured were 291-day Mn^{54}, 2.6-year Na^{22}, and probably a mixture of Sc^{46} and $Co^{56 + 58}$ (half-lives approximately 80 days); these nuclides were detected in relatively recent falls (the youngest, the Harleton, Texas, chondrite, was measured 21 days after fall). Furthermore, thorium was detected and measured in 5 of the 9 achondrites.

Of the 6 siderites studied, only Aroos and Sikhote-Alin showed detectable gamma radioactivity. In Aroos, measured 120 days after fall, Mn^{54} was the predominant radioactivity; Co^{60} was the only radioactivity measured in the 4 samples of Sikhote-Alin. In the other 4 old siderites, with a limit of detection of about 10 gammas/min/kg, no gamma-ray activity in the energy range 0.2 to 2 Mev was detectable.

Quantitative data on radioactive concentration are presented.

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CHAPTER 1

INTRODUCTION

The history and the composition of the solar system have fascinated scientists for centuries. Much attention has recently been directed to the meteorites, for they have proved to hold many clues to the solution of these questions. Meteorites are broadly classified according to the relative amounts of iron-nickel alloy and stony matter, as follows (1):

I Siderites (irons), consisting mainly of nickeliferous iron.

II Siderolites (stony irons), in which iron and stony matter are both present in large amounts.

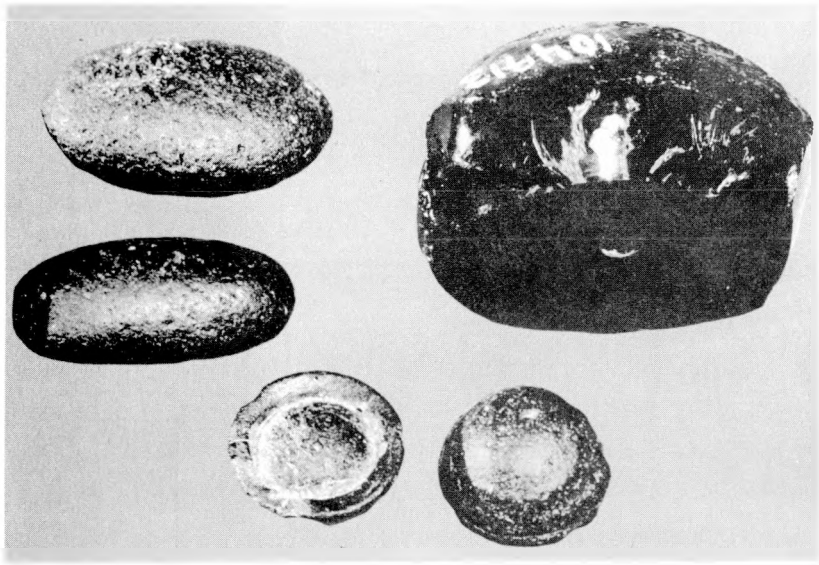
III Aerolites (stones), consisting mainly of stony matter with nickeliferous iron and troilite, when present, scattered through as small grains.

This last group subdivides into chondrites and achondrites. Chondrites contain chondrules, which are small silicate

bodies ranging from microscopic to, in rare instances, the size of a marble, with the vast majority being less than 2 mm in diameter. They are predominantly spheroidal in form. In some cases, they have suffered distortion since their formation, having been flattened or elongated (2). Achondrites contain no chondrules.

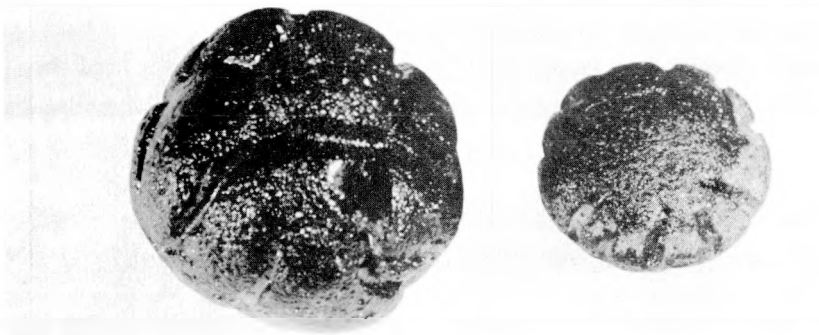
Usually included in a discussion of meteorites are the tektites, although their origin is uncertain. The tektites are rather strange looking silica glass objects (Fig. 1), which are found in extensive but limited areas of the earth and which have no connection with known volcanic regions. They are distinctly different in physical shape, in physical appearance, and in chemical composition (3) from natural glasses of known origin (i.e., obsidian and volcanic glasses). Some have very regular rotational forms (dumbbells, ellipsoidal shapes, etc.), some show interesting surface etching and marking, and many are the result of fragmentation. Their origin is controversial; some students of the subject favor a terrestrial and some an extraterrestrial origin (4).

Most scientists agree that meteorites are a part of the solar system. This is thought to be true, since practically all sporadic meteors and, hence the reasoning goes, most meteorites have elliptical orbits. LaPaz (5) argues that Watson (6) was incorrect in his statement that not one of



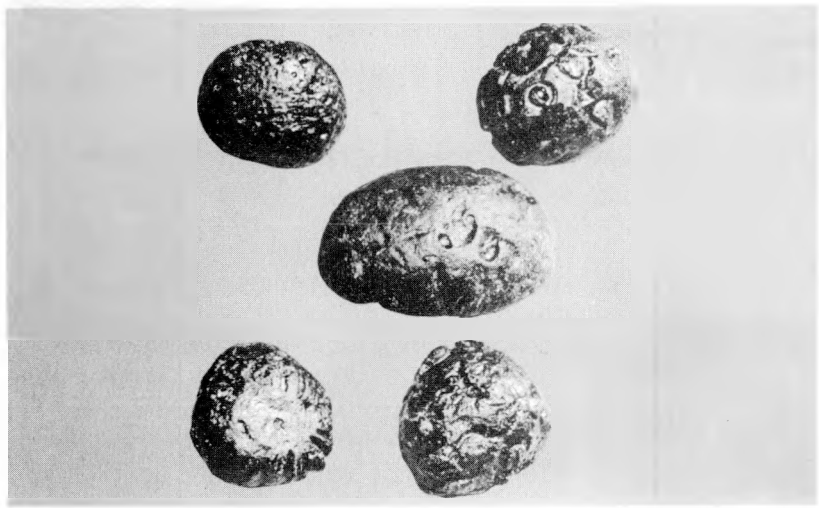
TEKTITES
(AUSTRALIA)

1 cm



TEKTITES
(COCO GROVE, P.I.)

1 cm



TEKTITES
(SANTIAGO, P.I.)

1 cm

Fig. 1. Tektites from Australia and Philippine Islands.

the sporadic meteors photographed in the Harvard Meteor Program had a hyperbolic orbit. (Upon coming close to the sun, meteorites of interstellar origin would move with respect to it on a hyperbolic orbit and thus would leave from the solar system to return to interstellar space.) However, there appears to be no question to Watson's statement that "if any meteors have hyperbolic velocities, they are less than 1 per cent of the total." It is generally concluded then that meteorites are a part of our solar system. They are also the only nonterrestrial bits of matter which are available to scientists for study in mass and, for this reason, are invaluable.

Since the meteorites are of such interest, it is obvious that accurate methods of analysis are of considerable importance. Because of the scarcity of meteorites for study and the desirability of conservation of samples, the need for nondestructive tests is realized. A nondestructive test system which averages over a fairly large sample is desirable to make sampling errors minimal.

The gamma radioactivity present in meteorites offers a unique method of meeting these requirements. By placing the specimen in a very sensitive gamma-ray spectrometer, it is possible to identify and to determine quantitatively those radionuclides which emit gamma rays. No treatment of the

meteorite is required, and it is not subjected to any artificial activation. The measurement can be made without removing the specimen from a protective plastic bag, if desired. Because of the unique penetrating nature of gamma rays, they reach the detector from the entire mass of the meteorite and not merely from a limited surface layer, so that the resultant measurement is characteristic of the sample as a whole.

Van Dilla, Arnold, and Anderson (7) have used a large sodium iodide crystal spectrometer to measure the radioactivity of meteorites. The results of their method were chiefly qualitative. Their work on meteorites was done as a preparative step for a project involving gamma-ray spectroscopy of the lunar surface.

The further development of the above technique to yield quantitative results seemed desirable in view of its advantages as listed in their paper. It was also thought that a more extensive set of measurements would be worthwhile, both to demonstrate the precision and accuracy obtainable and to supply additional data on the abundance of cosmic-ray-induced activities in representative meteorites. These are the objectives of this paper.

CHAPTER 2

THEORETICAL BACKGROUND

2.1 Source of Radionuclides in Meteorites

2.1.1 Natural Radionuclides

Primary natural radionuclides are unstable nuclides occurring at the present time because of their very long half-lives, so that detectable amounts have survived the time interval between the formation of the elements and the present time. Since there is evidence for the formation of the elements being about 6 aeons (1 aeon = 10^9 years) ago, a nuclide must have a half-life of at least $\sim 10^8$ years in order to have survived almost complete decay. Nuclides with half-lives of over $\sim 10^{10}$ years will have suffered little decay in this period. The most significant radiation sources are those with half-lives which are similar to the elapsed time, especially K^{40} ($T_{1/2}$ = 1.27 x 10^9 years), U^{238} ($T_{1/2}$ = 4.51 x 10^9 years), and Th^{232} ($T_{1/2}$ = 14.2 x 10^9 years) (Ref. 8). Of considerable importance with respect to U^{238} and Th^{232}

are the secondary natural radionuclides; these are relatively short-lived nuclides that occur in nature as a result of continual formation by decay of the primary natural radionuclides. Many of these emit gamma rays. All these naturally occurring radionuclides are of importance, because they are the major source of heat in planetary bodies, and they are extremely useful in dating methods. Since the meteorites are thought to be from a less differentiated system than the earth and, therefore, nearer to the primordial material, the contents of these heat-producing elements in the meteorites are of utmost importance. MacDonald (9) was also able to show with heat-flow measurements on the earth that the present rate of heat production in the earth is consistent with the supposition that the earth as a whole, or perhaps a major portion of it, is made up of chondritic material. The data do not allow distinction between an earth with an iron core and chondritic material for the rest or an entirely chondritic model. In either case, chemical differentiation of the mantle must be assumed, whereby the heat-producing elements are concentrated in the upper 600 to 700 km of the mantle. In connection with these calculations, the most important parameters are the contents of potassium, uranium, and thorium.

If meteorites are a part of the solar system, then they were probably formed at the same time as the rest of the

system, and the "age" of the meteorites (time elapsed since last total melting) should provide a lower limit on the age of our solar system. This "age" can be measured by counting the few atoms of the noble gases, helium and argon, that are trapped in the crystal lattices of the meteorites. In these methods, the noble gases are produced in the meteorite by decay of the naturally radioactive parent (i.e., uranium, thorium, and potassium, respectively), the gases representing the by-products of these nuclear transformations. Since the decay rates are known and are immutable, time is measured by comparing the relative abundances of the parent elements and their noble gas daughters present in a sample (assuming no diffusion losses). There are other "clocks" to tell geologic time -- the oldest, the lead/uranium method which, of course, also depends on the uranium content of the meteorite being dated.

2.1.2 Cosmic-Ray-Produced Radionuclides

Cosmic rays impinging on meteorites cause some nuclear reactions which can be used to yield another sort of "clock." By utilizing this phenomenon, it is possible to tell how long a meteorite has been exposed to cosmic radiation before reaching the earth. These cosmic ray "ages" are some tens of millions of years for stone meteorites (10).

Also of great importance is the information that can be gained on the cosmic-ray flux as a function of time and distance from the sun. Especially important is the variation of cosmic-ray intensity over long periods of time. Presently, only meteorite data are available for these studies. Specific cosmic-ray reactions studied in this paper are spallation and neutron-capture. These reactions, along with the specific nuclides formed, are discussed in the following sections.

2.1.2.1 Spallation Reactions

While nuclear reactions of all kinds occur when cosmic rays interact with meteorites, the ones of most importance in this study are the "spallation" reactions. The primary cosmic-ray flux is chiefly made up of high energy protons in the Bev energy range. The interaction of these protons with matter gives rise to a number of "secondaries" which are mainly neutrons, protons, and $\pi^{\pm}$ mesons. These secondaries have an energy spectrum ranging from the energy of the primary protons on down. The particles of most importance in causing the nuclear spallation reactions in meteorites are neutrons of energies in the range around 100 Mev. The spallation reaction can be simply thought of as an incoming particle (the 100-Mev neutron) striking the nucleus of an atom and knocking chips off it. The most likely products from a given material

are the products with atomic mass just below that of the target. The most likely particle being chipped off is a neutron. Other particles which are charged (protons, alpha particles, etc.) may also be ejected. Such reactions may yield products which are radioactive as, indeed, the species reported herein were. The nuclides found in this study are described below.

Aluminum^{26}--The (n,2n) and (n,pn) reactions on stony meteoritic material have a rather high probability of occurring from spallation in the 100-Mev region. In stone meteorites, Al^{26} is produced by similar low-energy reactions. Probably the predominant reaction is $Si^{28}(n,p2n)Al^{26}$ with some contribution from $Al^{27}(n,2n)Al^{26}$.

Sodium^{22}--Sodium^{22} is thought to be produced from similar low-energy reactions. The major contribution, in this case, is probably the spallation reaction on magnesium [namely, $Mg^{24}(n,p2n)Na^{22}$].

Manganese^{54}--This nuclide is also thought to be produced from 100-Mev or so neutron spallation; however, the reaction, in this case, is on iron. The iron content of chondrites is about 20 to 25 per cent by weight. Since stable Mn^{54} is rare in stones [MnO $\sim$ 0.3 per cent (11)], its spallation to Mn^{54} would not make a large contribution when compared with iron; therefore, the production of Mn^{54} in stones and irons would

be expected to occur by the same process. These nuclides were the predominant cosmic-ray-produced radioactivities in the stone meteorites. In addition, Mn^{54} was the predominant radioactivity detected in the Aroos siderite.* The relatively short half-lives of the Mn^{54} (291 days) and Na^{22} (2.6 years) precluded their measurement in a majority of meteorites; only recent falls will be above our limit of detection.

Additional short-lived gamma-ray emitters produced by cosmic-ray bombardment which were suggested by the gamma-ray spectrum of the Harleton chondrite were Sc^{46} (82.9 days), Co^{56} (77.2 days), and Co^{58} (71.3 days). The Co^{56} and Co^{58} were probably produced from spallation reactions on nickel [$Ni^{58}(n,p)Co^{58}$, for example]. Some contribution is probably furnished from spallation on cobalt as well. Turkevich (12) suggested that π^- absorption in the iron and nickel isotopes may contribute manganese, chromium, and cobalt nuclides in meteorites. Scandium^{46} is produced principally from spallation on iron. These nuclides are made practically in the same way in stones and irons, because of the low abundance of elements between iron and calcium in the stones.

The principal activities found in the siderites were the aforementioned Mn^{54}, detected in Aroos, and Co^{60}, found in Sikhote-Alin.

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*For acknowledgments of source of meteorites, see page 95.

2.1.2.2 Neutron-Capture Reaction

Unlike the species discussed above, the production of Co^{60} in the Sikhote-Alin siderite was found to be more than a factor of 10 over what would be expected from spallation. The principal reaction producing Co^{60} in the Sikhote-Alin was neutron-capture by Co^{59}. It should be noted that the Aroos siderite (measured at 120 days after fall), which was more recent than Sikhote-Alin (measured ~13 years after fall), had a Co^{60} content which was down more than an order of magnitude below Sikhote-Alin. This is because of the mechanism for production of radionuclides by neutron-capture. In order that a neutron be captured, it must be moderated which, in a high mass number material like iron, requires many collisions and hence large amounts of material. Van Dilla, Arnold, and Anderson (7) estimated that a mass of several tons was necessary for an optimum flux of slow neutrons in a siderite.

2.2 Some Aspects of Gamma Radiation

2.2.1 Properties of Gamma Radiation

Gamma rays are electromagnetic radiation of a rather penetrating nature. There are three mechanisms by which gamma rays can be absorbed.

The first case, which is predominant at low energies, is the photoelectric effect. Figure 2 shows this process schematically. In this process, the gamma ray of energy $h\nu$ ejects a bound electron from an atom and imparts to it an energy equal to $h\nu - b$, where b is the binding energy of the electron. The quantum of radiation disappears completely in this process. The photoelectric absorption is approximately proportional to Z^5 (Z is the atomic number of the absorber). The photoelectric effect is frequently used to determine gamma-ray energies.

Instead of transmitting its entire energy to the electron, a gamma ray may lose only part of its energy to an electron, in which case the gamma ray will be deflected from its original path and will be degraded in energy. This process is known as the Compton effect or Compton scattering and is shown schematically in Fig. 3. Compton scattering per electron is independent of Z and, therefore, the scattering coefficient per atom is proportional to Z. Compton scattering is the predominant process in the energy range 0.2 to 3 Mev, which was the range investigated in this report.

The third mechanism by which gamma rays may be absorbed is the pair-production process (Fig. 4). An energy of the incident gamma ray of >1.02 Mev is necessary for this process to occur. Pair-production is unimportant except at even

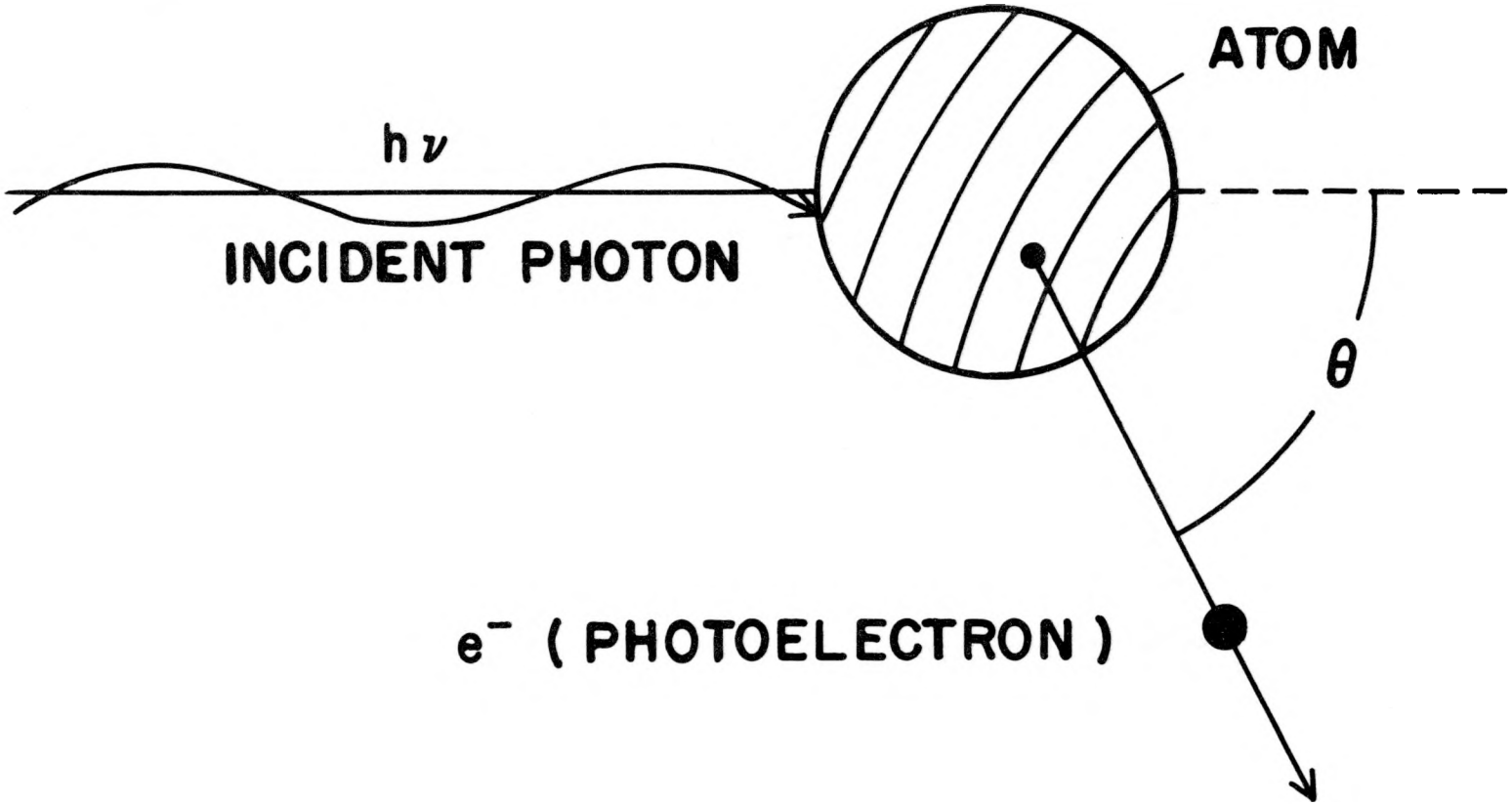

Fig. 2. Schematic diagram of the photoelectric effect (13).

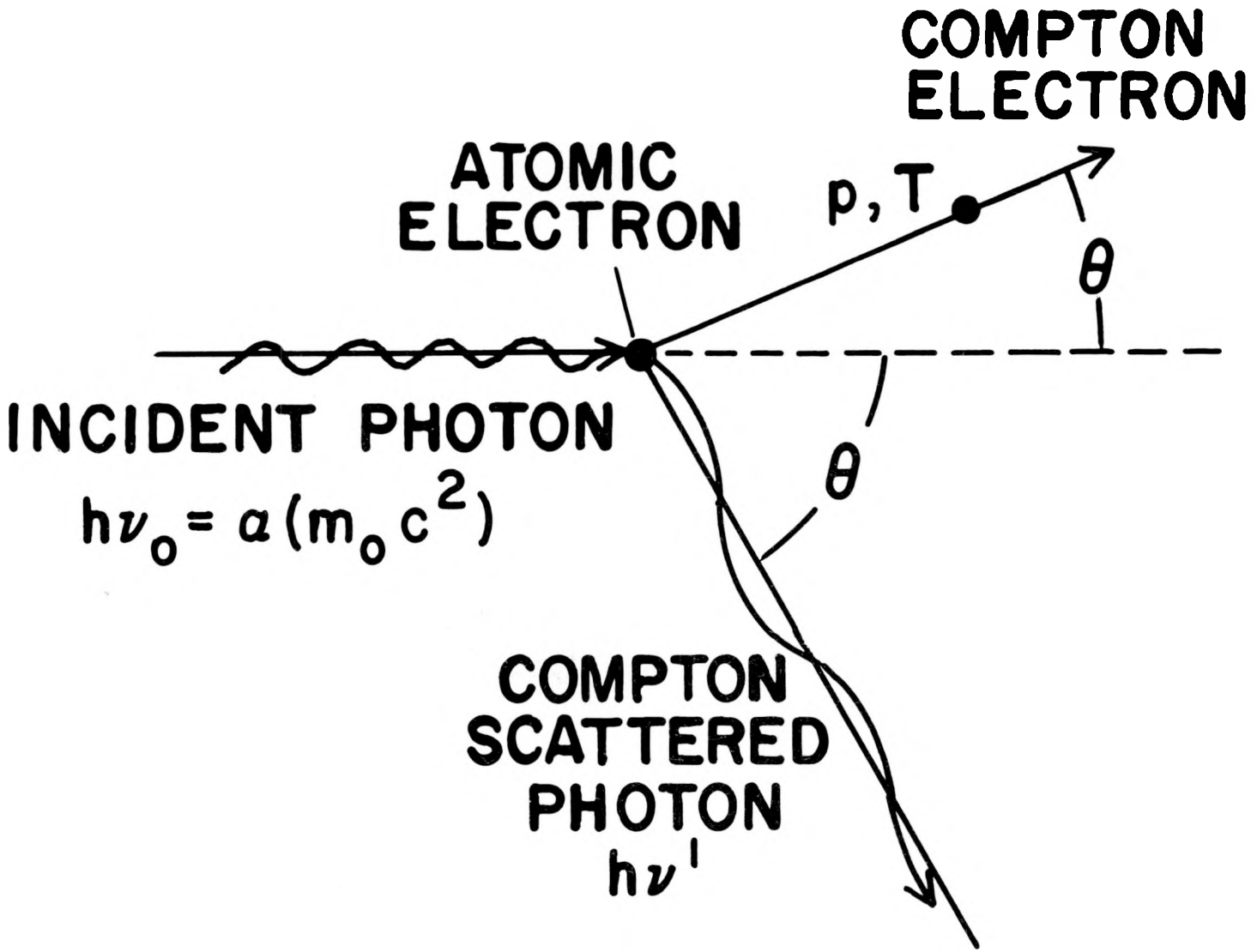

Fig. 3. Schematic diagram of Compton scattering (13).

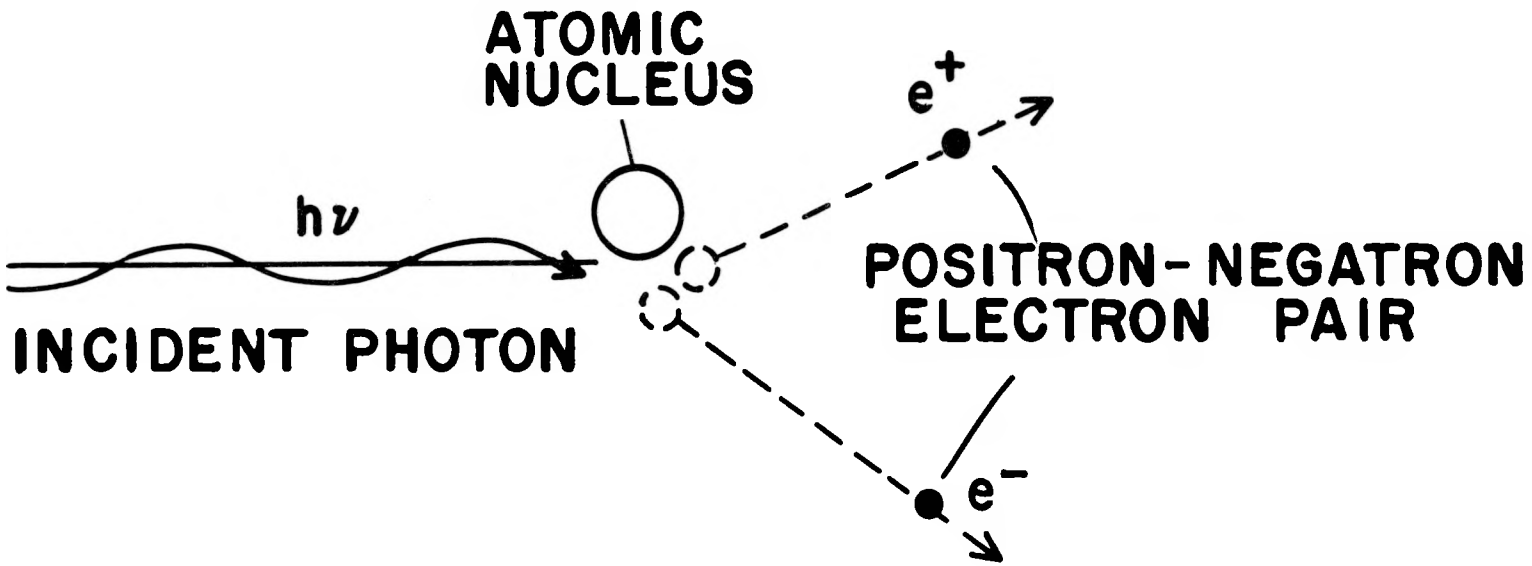

Fig. 4. Schematic diagram of the pair-production effect (13).

higher energies (>3 Mev). Pair-production is always followed by the annihilation of the positron yielding two 0.51-Mev photons.

2.2.2 Gamma-Ray Emission

A nucleus which, for some reason or another, is in an excited state may return to a more stable ground state by giving up its excitation energy. This may be accomplished in a variety of ways, the most common of which is by the emission of electromagnetic radiation. This radiation is termed gamma radiation and has an energy determined by its frequency by the relationship $E = h\nu$, where E is the energy, h is Planck's constant, and ν is the frequency of the radiation. Any event which leaves a nucleus in an excited state (many alpha and beta decay processes) is usually followed by gamma radiation. Gamma rays of energies from 10 kev to about 7 Mev have been observed in radioactive processes. Gamma-ray emission may be accompanied by or replaced by another process: the emission of internal-conversion electrons. Internal-conversion can be pictured as being a process where the emitted gamma ray has undergone photoelectric absorption by an orbital electron, the energy of the ejected electron being the difference in the gamma-ray energy and the binding energy of the electron in the atom. In this case, the only electromagnetic radiation emitted will be X rays.

2.2.3 Principles of Detection of Gamma Rays

The detection of gamma rays depends upon one of the three processes of absorption listed above. Specifically, with a NaI (Tl) crystal, an interaction of the gamma rays with the crystal causes "scintillations" (i.e., the emission of visible light in an amount proportional to the energy of the ejected electron). The high Z of the iodine (desirable for production of photoelectric effect) and the high density of the sodium iodide crystals make them especially suitable for gamma-ray measurements. Typically, the crystal is fixed to the photosensitive face of one (or several) multiplier phototubes with silicone oil, to provide a good optical contact at the interface. A light-tight enclosure with a good reflector (MgO was used for the LASL crystals) is essential for light collection and hence resolution. The sodium iodide crystals have a very high light yield. The crystal must be large enough to contain a sufficient number of the Compton-scattered gamma rays produced in the crystal itself so that the resulting pulses will correspond to the total gamma-ray energy, since the energy used to overcome the electron's binding energy will also contribute to the scintillation through

X-ray and Auger-electron* emission. In addition to this "photopeak," there will be a continuous distribution of smaller energies corresponding to the Compton electrons. This continuum will depend on the size of the crystal to the extent that the crystal is not large enough to contain the entire sequence of processes that consume the initial gamma-ray energy. Gamma-ray spectrometry based on scintillation response offers the best sensitivity but less than the best resolution. The resolution is the width of the peak at half-height divided by the energy of the peak. Figure 5 shows a Cs^{137} spectrum on the Los Alamos 7-1/2 x 4-in. NaI (Tl) spectrometer and illustrates the resolution, which places the limit on the number of gamma-ray emitters that can be detected simultaneously.

The difficulty in obtaining quantitative results from gamma-ray spectra is that the efficiency of the instrument is dependent on a number of factors which must be taken into

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*Auger electron: when an electron is knocked from its shell, a vacancy is created. Especially for the emission of a K electron from an atom of high Z, the resulting excitation is appreciable. The vacancy is usually filled by an electron from the next higher shell. If an L electron falls into the K shell, the difference between the K and L binding energies may be emitted as a characteristic X ray or may be used in an internal photoelectric process analogous to internal-conversion. In the latter case, an additional extranuclear electron from the L or M, etc., shell is emitted with a kinetic energy equal to the characteristic X-ray energy minus its own binding energy. Such electrons are called Auger electrons.

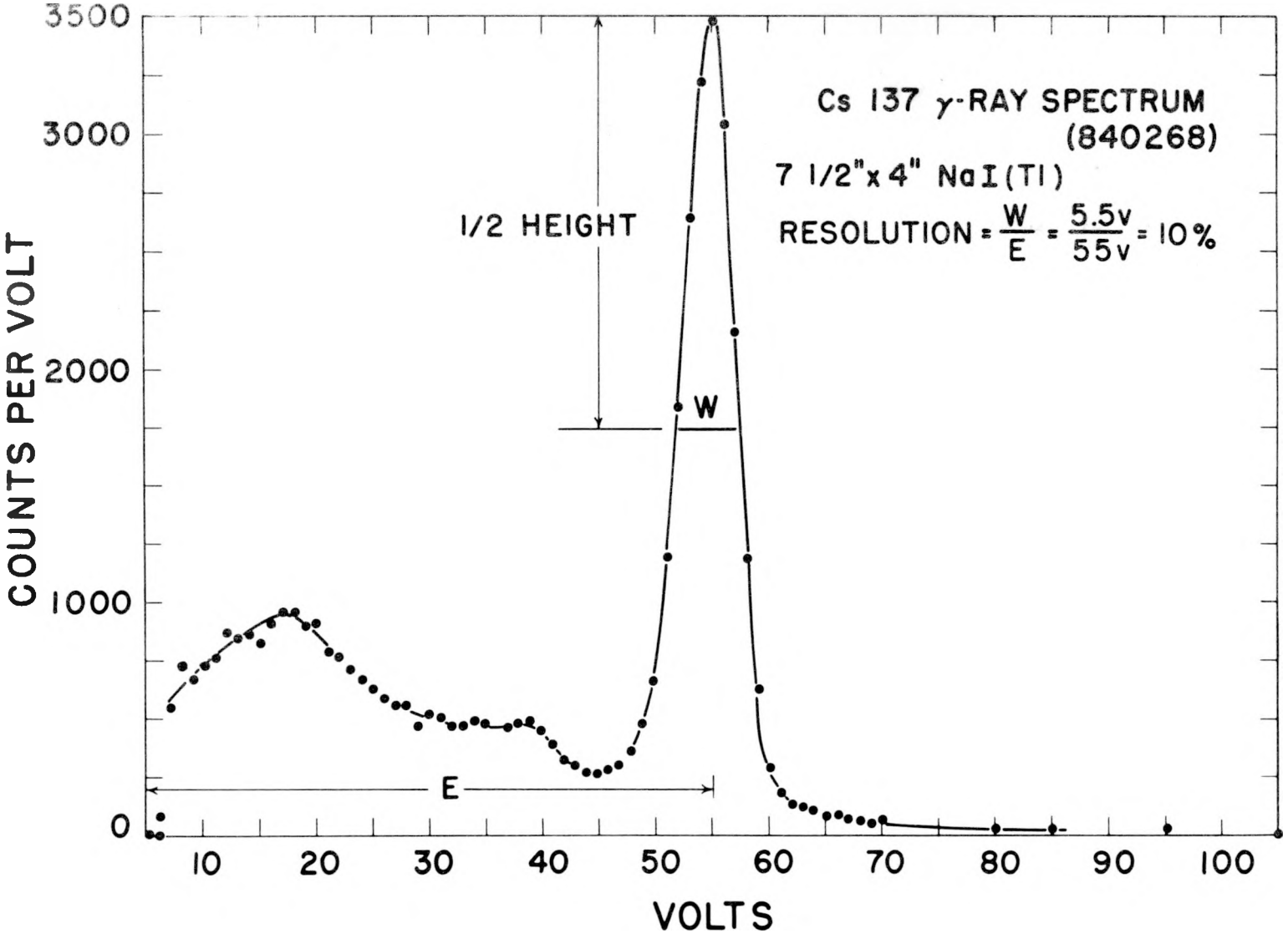

Fig. 5. Resolution of a Cs^{137} spectrum on a 7-1/2 x 4-in. NaI (Tl) spectrometer.

account. This amounts to calibrating the crystal-sample combination for the following factors:

a. Geometrical efficiency (i.e., what fraction of the gamma rays leaving the sample pass through the crystal).

b. Inherent crystal efficiency and photofraction as a function of the gamma-ray energy.

c. Crystal resolution as a function of energy.

d. Self-absorption of gamma rays in the meteorite material and in the mockup.

e. Gamma emission rate of the nuclear decay being measured.

Thus, a given calibration is specific for a given crystal-sample-nuclide combination. Taking the above factors in order, it will be shown how the problems can be solved by making a model of the meteorite which is very similar to it with respect to geometry, gamma-ray self-absorption, and scattering effects. An accurately known amount of some radioactivity with a gamma-ray energy spectrum similar to the unknown is necessary.

a. The geometrical efficiency is dependent only on the shape of the object and the distribution of the nuclide in question. Since the radioactivities measured were, for all practical purposes, uniformly distributed in both the meteorite

and the mockup, the distribution effect was eliminated. The model was made the same shape as the meteorite, which eliminated the factor of geometrical corrections entirely.

b. A correction taking into account the inherent crystal efficiency and photofraction as a function of gamma-ray energy became necessary because a gamma ray of one energy was used to calibrate the gamma ray of another energy [i.e., the K^{40} (1.46-Mev gamma ray) was used to calibrate the Al^{26} (1.83-Mev gamma ray, etc.)]. The correction was necessary since two sources with identical activity but different gamma-ray energies will give different counting rates in the photopeak. The corrections used to account for this effect were taken from the Monte Carlo calculations of Miller, Reynolds, and Snow (14) for a broad parallel beam using an 8 x 4-in. NaI (Tl) crystal. That this is a sufficient correction is shown by calculating the Al^{26} content of several meteorites by this method and comparing the results with the Al^{26} content calculated by comparison with an Al^{26} mockup. This was done and the agreement was satisfactory.

c. The resolution differences between the different energies were corrected for as follows. Photopeak band widths were chosen so that the counts within a certain energy band could be compared directly with a different energy band. In

order to do this, the resolution at two energies was measured: R (0.66 Mev) = 9.6 per cent, and R (1.46 Mev) = 6.8 per cent, where R = resolution. Knowing that $R^2 = \alpha + \frac{\beta}{E}$ (15), where α and β are constants characteristic of a particular crystal, and E is the energy of the gamma ray in Mev, two equations can be set up which can be solved simultaneously for α and β. Hence,

$$(9.6)^2 = \alpha + \frac{\beta}{0.66}$$

$$(6.8)^2 = \alpha + \frac{\beta}{1.46}$$

$$\alpha = 8.5 \text{ and } \beta = 55.2$$

Thus, for the Los Alamos 7-1/2 x 4-in. NaI (Tl) crystal,

$$R = 8.5 + \frac{55.2}{E}$$

The resolution at 1.83 Mev is, therefore, calculated to be 6.25 per cent. (A measured value taken later was 6.22 per cent.) The measured resolution is given by $R = \frac{W}{E}$, where W is the width of the photopeak at half of the maximum height, and E and R are the same as before. Rearranging and substituting 1.46 and 1.83 Mev, the desired ratio of the widths of the photopeaks is seen to be:

$$\frac{W_{1.46}}{W_{1.83}} = \frac{6.8 \times 1.46}{6.25 \times 1.83} = 0.87$$

The K^{40} band width was chosen from 1.36 to 1.56 Mev. This choice was made by considering two conflicting parameters, namely, the desire to have a wide energy band to decrease the uncertainty from counting statistics versus the desire to keep the energy band narrow so that advantage was taken of the resolution. The narrow band was desirable to keep interference at a minimum from gamma rays of energy near to that being quantitated. The decision was to include essentially all of the photopeak but none of the Compton continuum. A band width was chosen by assuming that a photopeak can be represented by a normal distribution curve. Since the entire photopeak area was wanted, the width which included 98 per cent of the area was taken from the "Normal Curve of Error" table in the Handbook of Chemistry and Physics. This width was seen to be $\pm$ 2.34 σ, which is twice the width at half maximum height or twice the resolution width (13). Therefore, using twice the resolution width, the energy band chosen for K^{40},

2 (6.8 per cent) (1.46 Mev) = 0.20 Mev,

centered on 1.46 Mev. Similarly for Al^{26}, the width chosen was:

$$2\ (6.25\ \text{per cent})\ (1.83\ \text{Mev}) = 0.23\ \text{Mev}.$$

This was rounded to 0.24 Mev in order that full channel widths would be used. The Al^{26} band was centered on 1.82 Mev. This meant then that the ratio of the band widths actually chosen was:

$$\frac{W_{1.46\ \text{Mev}}}{W_{1.83\ \text{Mev}}} = \frac{0.20\ \text{Mev}}{0.24\ \text{Mev}} = 0.835,$$

compared with the desired 0.87. This is as close as one can get using full 20-kev channels.

d. The self-absorption of the 1.46-Mev K^{40} gamma ray and the 1.83-Mev Al^{26} gamma ray in the meteorite was calculated using the following equation:

$$\text{Absorption} = e^{-(\mu_o/p)(px)}$$

where e = base Naperian logarithms; μ_o = linear absorption coefficient; p = density; and x = thickness of the absorber. The term μ_o/p used was that in aluminum (13): μ_o/p (aluminum, 1.46 Mev) = 0.050 and μ_o/p (aluminum, 1.83 Mev) = 0.045. Average meteorite density was assumed to be 3.5 g/cc. The average thickness of the meteorite was approximated. That

these approximations are not very critical can be shown by the following. If

$$t = 0.5 \text{ in.}, \text{ correction} = 0.978$$

$$t = 1.0 \text{ in.}, \text{ correction} = 0.955$$

$$t = 1.5 \text{ in.}, \text{ correction} = 0.933$$

Therefore, by an error of a factor of 3 in thickness, one gets a discrepancy of only 4.5 per cent in this correction factor.

The mockup material was a mixture of iron-powder and NaCl (or KCl) which had an electron density* close to that of the stone meteorite.

e. The gamma emission rate per disintegration is not a constant among the various nuclides. An examination of the decay schemes of the various nuclides investigated (i.e., K^{40}, Al^{26}, Mn^{54}, Na^{22}, Co^{56+58}, Sc^{46}, and Co^{60}) was, therefore, necessary. The decay scheme of Al^{26} (16,17) is shown as an example in Fig. 6. A complete decay scheme includes all the modes of decay of the nuclide, their abundances, the energies of the radiations, the sequence in which the radiations are emitted, and the measurable half-lives of any

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*Electron density of an element is given by $\frac{Z}{A}N$ electrons/g, where Z is the atomic number, A is the atomic weight, and N is Avogadro's number.

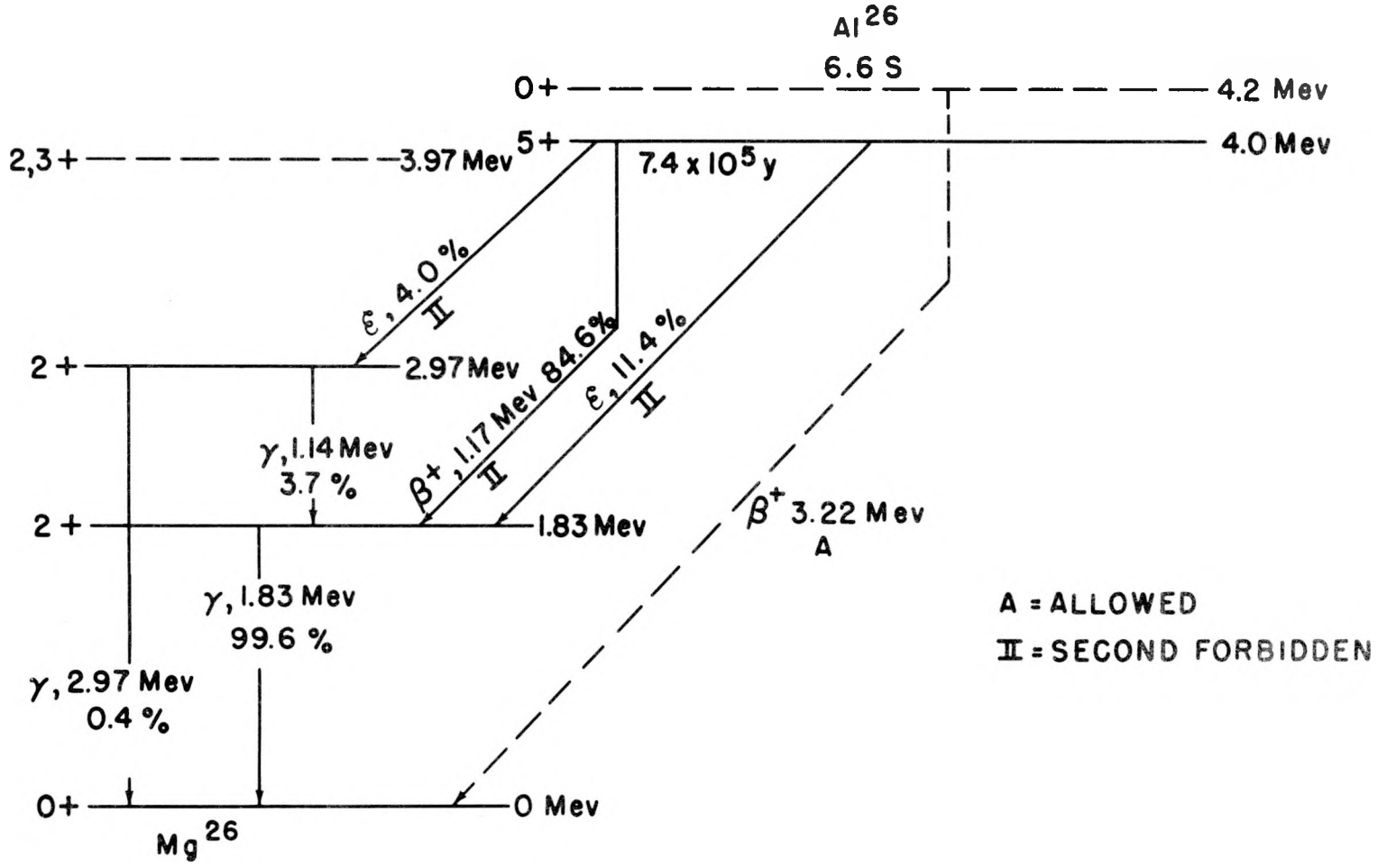

Fig. 6. Decay scheme of Al^{26} (16,17).

intermediate states. Where possible, spin and parity assignments of the various energy levels involved are included in the decay scheme. The amount of detail known about a given decay scheme depends, of course, very strongly on the refinements in instrumentation and techniques.

From the Al^{26} decay scheme shown in Fig. 6, the following information is available. The half-life on the long-lived isomer of Al^{26} is 7.4×10^5 years. $Aluminum^{26}$ has three modes of decay: one branch (11.4 per cent), which goes to the 1.83-Mev energy level by electron capture; the second branch (84.6 per cent by positron emission), which also goes to the 1.83-Mev energy level; and the third branch, which is another electron capture (4.0 per cent), which goes to a 2.97-Mev energy level. The 2.97-Mev level then goes to the 1.83-Mev 3.7 per cent with emission of a 1.14-Mev gamma ray. A 0.4 per cent branch gives off a 2.97-Mev gamma ray to go to the ground state. This leaves a total of 99.6 per cent of the disintegrations at the 1.83-Mev energy level. This level decays by gamma-ray emission to the ground state. If the 0.51-Mev positron annihilation photon is used for calibration purposes, one would notice that the positron decay occurs only 84.6 per cent of the disintegrations.

The experimental data obtained by the above explained technique are reported in the next two chapters.

CHAPTER 3

STONE METEORITES

The stones are the most abundant group of the meteorites, making up about 90 per cent of the seen falls. Of these, the chondrites are the most abundant, the achondrites accounting for only about 10 per cent of the total of 628. Only 19 carbonaceous chondrites are known, and all these were seen to fall (1). A representative sampling was desired, and a total of 32 samples of 25 chondrites, 4 carbonaceous chondrites, and 9 achondrites have been measured. Potassium^{40} and Al^{26} were the predominant radioactivities detected; more unusual gamma-ray emitters detected were 291-day Mn^{54}, 2.6-year Na^{22}, and probably a mixture of Sc^{46} and Co$^{56 + 58}$ (all half-times approximately 80 days). These nuclides were detected as a result of measurement of a few relatively recent falls (the youngest, the Harleton, Texas, chondrite was measured 21 days after fall). Furthermore, thorium was detected in 5 of the 9 achondrites, with a suggestion of some uranium in these cases.

3.1 Experimental Procedure

All measurements taken in this study were made with large (7-1/2 x 4-in. and 9-1/2 x 6-in.) NaI (Tl) crystal spectrometers. These detectors have been described in detail by Van Dilla (18). A major problem encountered in interpreting quantitatively the gamma-ray spectrum of an irregularly shaped meteorite consists of accounting for self-absorption and geometry effects. The problem has been solved by making a thin hard shell the shape of the meteorite (as shown in Fig. 7). First, the meteorite is covered with aluminum foil, the foil being pressed tightly onto the surface of the meteorite to conform closely to its shape. A molding compound, Rezolin Epoxy F,* is then painted on the aluminum-coated meteorite in two halves with an unpainted strip about 1/16 in. wide around the middle. After the compound hardens, the two halves are pulled apart from the meteorite and then "glued" together with more molding compound to form the completed shell. Iron-powder thoroughly mixed with a known amount of KCl is packed into the shell so that the total weight of the mockup (including the shell) is equal to the weight of the meteorite. Iron-powder was chosen as the major constituent of the shell filling because its interactions with gamma rays

*Rezolin L-933A Epoxy Resin "F," Rezolin, Inc., Santa Monica, California.

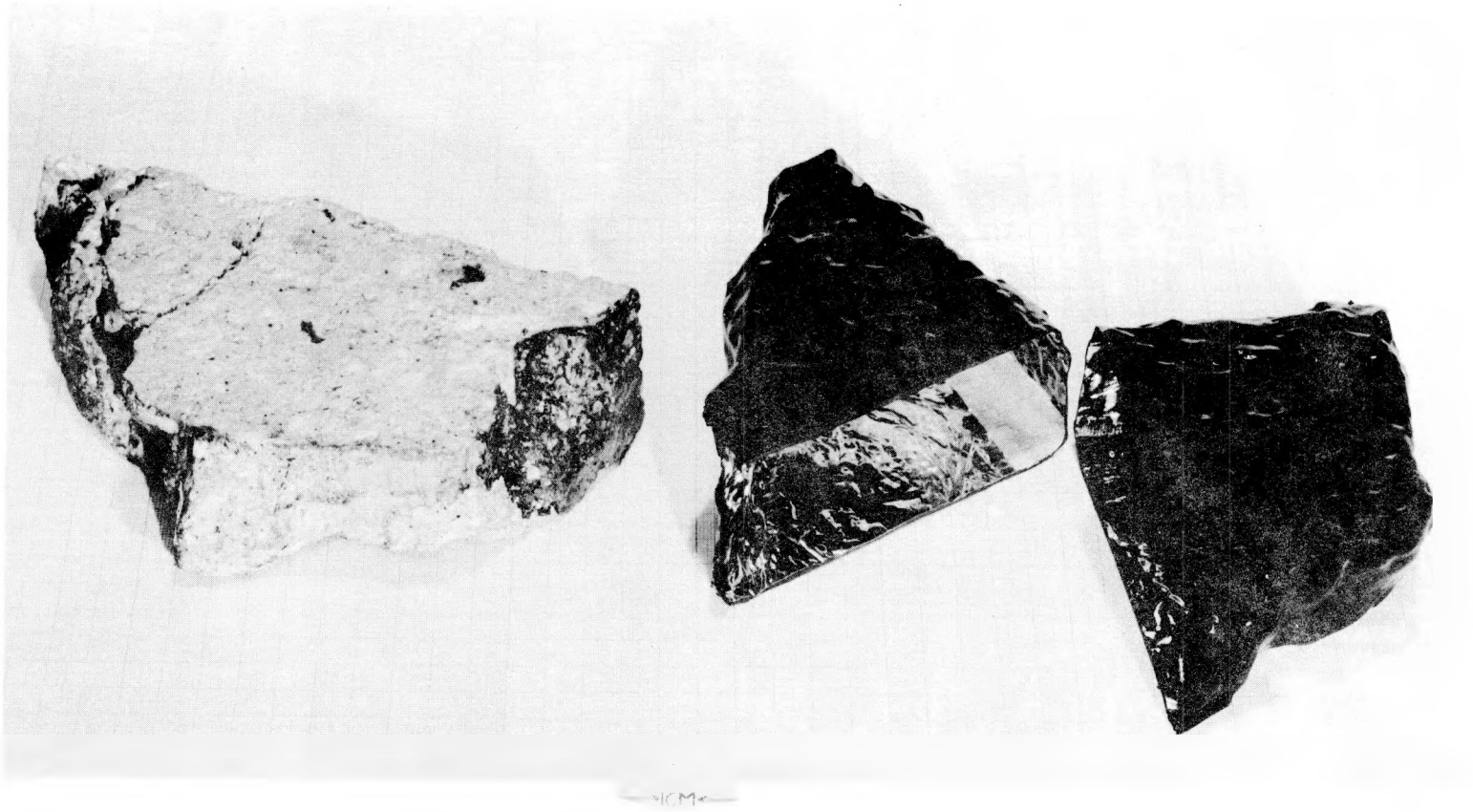

Fig. 7. Norton County achondrite and Rezolin shell.

in the region of 0.5 to 3 Mev are very similar to those of chondrites. At this energy, Compton absorption and scattering are virtually the only interactions taking place in the chondrite. Iron-powder has an electron density close to that of the average chondrite, and its atomic number, although larger than for chondrites, is still low enough to make pair-production and photoelectric effects negligible. The average chondritic composition used was based on that given by Urey (11), neglecting constituents of less than 1 per cent (shown in Table 1). On this basis, the chondritic electron density is 2.92×10^{23} electrons/g. This value is not very sensitive to changes in chemical composition because of the very similar electron density of the individual components. Variation in the free iron will produce the greatest change but, even in this case, doubling the average amount and making the corresponding decrease in silica content will change the average electron density by only about 2 per cent. The electron densities of the mockups ranged from 2.84 to 2.89×10^{23} electrons/g.

After counting each mockup in the same geometry as the meteorite, direct comparison of the K^{40} photopeak areas yielded the meteorite potassium content quantitatively. The Al^{26} content was then calculated by comparing the 1.83-Mev Al^{26} photopeak with the known 1.46-Mev K^{40} photopeak of the

TABLE 1. AVERAGE CHONDRITIC COMPOSITION USED IN ELECTRON DENSITY CALCULATIONS (BASED ON UREY)

Component	Average Chondrite (per cent composition)	Electron Density x 10^{23} electrons/g
SiO_2	38.04	3.00
MgO	22.84	3.00
FeO	12.45	2.85
Al_2O_3	2.50	2.98
CaO	1.95	3.01
Fe	11.76	2.80
FeS	5.73	2.88
Ni	1.34	2.88
Total	96.61	

mockup and making small corrections for efficiency, photofraction, peak width, and self-absorption, as explained previously.

For the special case of the more recent falls, mockups were made for all the predominant activities present (namely, Al^{26}, K^{40}, Na^{22}, and Mn^{54}). Thorium mockups were made for comparison with the 5 achondrites having this activity. An attempt was made to quantitate the uranium present, if any, but the results were inconclusive. The presence of uranium could have been confirmed and the amount quantitated by use of two crystals in coincidence. A spectrometer of this type is in operation at Los Alamos and uses two matched 8 x 4-in. NaI (Tl) crystals (19). However, lack of time prevented carrying out this investigation. The nature of the standards used to calibrate the unknown activities in the stones is as follows:

Potassium^{40}--An accurately weighed amount of KCl constituted the potassium standard.

Sodium^{22} and Manganese^{54}--Both the Na^{22} and the Mn^{54} solutions were calibrated at the National Bureau of Standards with an accuracy estimated at $\pm$ 2 per cent.

Aluminum^{26}--The Al^{26} was calibrated by comparing the positron annihilation peak of Al^{26} with that of the National Bureau of Standards-calibrated Na^{22} and also by comparing

the 1.83-Mev peak of Al^{26} with a known 1.46-Mev K^{40} peak, and once more making the necessary small corrections for photofraction, peak width, and self-absorption. The two results were in fair agreement with each other.

Thorium--The thorium was calibrated by the New Brunswick Laboratory* with an accuracy stated at ± 2 per cent.

All other activities (including Al^{26}, in practically all cases) were compared with some other known gamma-ray photopeak in the meteorite, the known gamma-ray affording an internal calibration standard.

3.2 Results

The results on potassium and Al^{26} assays on stone meteorites are shown in Table 2, along with values obtained by other workers; agreement is good. The constancy of the Al^{26} contents for the chondrites provided an inner check against gross errors.

The results for K^{40}, Al^{26}, Mn^{54}, Na^{22}, and probably Sc^{46} plus $Co^{56 + 58}$ for Bruderheim, Harleton, Ehole, and Hamlet are shown in Table 3. It must be realized that although Ehole was considered to contain only potassium, Al^{26}, Na^{22},

*U. S. Atomic Energy Commission, New Brunswick, New Jersey.

TABLE 2. POTASSIUM AND ALUMINUM^{26} CONTENTS OF CHONDRITES

Meteorite	Weight (kg)	Per Cent Potassium by Weight		Aluminum^{26} (dis/min/kg)	
		This Paper	Other Reports	This Paper	Other Reports
CHONDRITES					
Abee	2.709	0.087 ± 0.004	---	51 ± 5	---
Achilles	0.675	0.070 ± 0.005	---	50 ± 5	50 ± 5[b]
Archie	3.773	0.076 ± 0.004	---	42 ± 4	---
Beardsley I	0.857	0.090 ± 0.009	0.0906 ± 0.0013[c]	50 ± 10	---
Beardsley II	0.637	0.125 ± 0.004 0.119 ± 0.004[a]	0.124[j]	55 ± 5	---
Beardsley III	4.280	0.105 ± 0.004	0.102 ± 0.003[d] 0.100 ± 0.002[e]	39 ± 4	---
Bruderheim	2.150	0.089 ± 0.002	0.0896 ± $^{0.0023}_{0.0046}$[f] 0.116 ± 0.008[g]	57 ± 2	60 ± 6[b]
Calliham	1.044	0.084 ± 0.004	---	53 ± 5	---
Cavour	4.090	0.071 ± 0.006	0.075[h] 0.070 ± 0.007[i]	41 ± 8	--- 50 ± 10[i]
Cherokee Springs I	2.682	0.078 ± 0.003	---	40 ± 3	---
Cherokee Springs II	5.023	0.077 ± 0.003	---	41 ± 3	---
Ehole	1.528	0.083 ± 0.003	---	33 ± 2	---
Forest City I	0.432	0.078 ± 0.006	0.084 ± 0.003[e]	34 ± 5	---
Forest City II	4.218	0.072 ± 0.004	0.082 ± 0.0014[c] 0.083 ± 0.003[k]	35 ± 4	---
Hamlet	0.730	0.096 ± 0.008	---	52 ± 7	---
Harleton	4.400	0.069 ± 0.003	---	43 ± 3	---

TABLE 2 (continued)

Meteorite	Weight (kg)	Per Cent Potassium by Weight		Aluminum^{26} (dis/min/kg)	
		This Paper	Other Reports	This Paper	Other Reports
Holbrook	1.121	0.083 ± 0.004	0.087 ± 0.003[d] 0.083[l]	58 ± 6	---
Ladder Creek I	1.010	0.086 ± 0.003	0.091[h]	32 ± 3 32 ± 3[i]	---
Ladder Creek II	4.048	0.088 ± 0.003	---	36 ± 3	---
La Lande	1.375	0.060 ± 0.004	0.075[h] 0.062 ± 0.006[i]	49 ± 5	---
Mocs	0.681	0.090 ± 0.004	0.090 ± 0.003[d] 0.087 ± 0.003[e]	52 ± 5	---
Modoc	1.172	0.088 ± 0.004	0.089 ± 0.003[m]	52 ± 5	---
Morland	0.750	0.085 ± 0.003	---	47 ± 5	---
Ness	0.872	0.080 ± 0.004	0.077 ± 0.003[d]	56 ± 6	---
Pantar	1.343	0.082 ± 0.004	0.088 ± 0.003[m]	53 ± 5	---
Plainview	3.450	0.084 ± 0.003	---	56 ± 5	54 ± 5[n] 68 ± 5[o] 38.5 ± 1.6[p]
Potter I	1.441	0.073 ± 0.003	0.079 ± 0.003[m]	54 ± 5	---
Potter II	1.030	0.069 ± 0.003	---	50 ± 5	---
Richardton I	0.620	0.084 ± 0.004	0.0818 ± 0.0012[c]	52 ± 6	63 ± 4[n]
Richardton II	5.679	0.069 ± 0.003	0.084 ± 0.003[d]	29 ± 3	65 ± 8[p]
St. Chinian	0.096	0.132 ± 0.066	---	52 ± 26	---
Sylacauga	1.682	0.068 ± 0.003	---	49 ± 5	---

TABLE 2 (continued)

Meteorite	Weight (kg)	Per Cent Potassium by Weight		$Aluminum^{26}$ (dis/min/kg)	
		This Paper	Other Reports	This Paper	Other Reports
CARBONACEOUS CHONDRITES					
Felix	1.247	0.037 ± 0.003	0.037 ± 0.003[d]	38 ± 4	---
Indarch	1.776	0.087 ± 0.003	0.088 ± 0.003[m]	40 ± 4	---
Mighei	0.750	0.038 ± 0.003	0.040 ± 0.003[d]	26 ± 3	---
Murray	1.243	0.045 ± 0.005	0.027 ± 0.003[d]	44 ± 4	---
			0.033		
ACHONDRITES					
Bishopville	0.583	0.126 ± 0.006	0.123 ± 0.004[m]	63 ± 6	---
			0.083 ± 0.003[m]		
Juvinas	0.833	0.048 ± 0.006	0.033 ± 0.003[d,m]	98 ± 12	---
Moore County	0.500	0.017 ± 0.007	0.021 ± 0.015[e]	55 ± 7	---
0.500 + 0.685		0.015 ± 0.003	0.0187 ± 0.0015[c]	55 ± 5	---
Norton County	1.590	0.012 ± 0.003	0.007 ± 0.002[e]	53 ± 5	---
			0.033[r]		
Nuevo Laredo	0.202	0.050 ± 0.015	0.0367 ± 0.0012[c]	60 ± 28	---
			0.044[q]		
			0.042 ± 0.003[m]		
Pasamonte	0.873	0.038 ± 0.003	0.036 ± 0.005[s]	73 ± 6	---
			0.043 ± 0.003[e]		
			0.0425 ± 0.0012[c]		
Peña Blanca	0.439	0 ± 0.004	---	48 ± 8	---
Sioux County	0.998	0.024 ± 0.004	0.0335 ± 0.0016[c]	90 ± 12	---
			0.033 ± 0.002[s]		
Stannern	1.133	0.076	0.060 ± 0.005[m]	96 ± 12	---

TABLE 2 (continued)

FOOTNOTES:

[a]This determination was run on a double 8 x 4-in. NaI (Tl) crystal setup.

[b]Honda, M., S. Umemoto, and J. R. Arnold, J. Geophys. Res. 66, 3541-3546 (1961).

[c]Gast, P. W., Geochim. Cosmochim. Acta 19, 1-4 (1960).

[d]Edwards, G., and H. C. Urey, Geochim. Cosmochim. Acta 7, 154-168 (1955).

[e]Geiss, J., and D. C. Hess, Astrophys. J. 127, 224-236 (1958).

[f]Reynolds, J. H., private communication with R. E. Folinsbee.

[g]Baadsgaard, H., F. A. Campbell, R. E. Folinsbee, and G. L. Cumming, J. Geophys. Res. 66, 3574-3577 (1961).

[h]Maynes, D., private communication.

[i]Van Dilla, M. A., private communication.

[j]Gast, P. W., National Academy of Sciences-National Research Council Publication 845 (1961), pp. 85-89.

[k]Folinsbee, R. E., J. Lipson, and J. H. Reynolds, Geochim. Cosmochim. Acta 10, 60-68 (1956).

[l]Mason, B., and H. B. Wiik, Geochim. Cosmochim. Acta 21, 276-283 (1961).

[m]Edwards, G., Geochim. Cosmochim. Acta 8, 285-294 (1955).

[n]Ehmann, W. D., and T. P. Kohman, Geochim. Cosmochim. Acta 14, 364-379 (1958).

[o]Anders, E., Geochim. Cosmochim. Acta 19, 53-62 (1960).

[p]Chakrabartty, M., Carnegie Institute of Technology Progress Report in Nuclear Chemistry (1960-1961), pp. 68-73.

[q]Reynolds, J. H., and J. I. Lipson, Geochim. Cosmochim. Acta 12, 330-336 (1957).

[r]Wiik, H. B., Geochim. Cosmochim. Acta 9, 279-289 (1956).

[s]Wänke, H., and H. König, Final Report, Max-Planck-Institut für Chemie, Mainz, Contract AF61(514)-1332 (1960).

TABLE 3. RADIOACTIVITY OF THE HARLETON, BRUDERHEIM, EHOLE, AND HAMLET CHONDRITES

	Harleton*	Bruderheim*	Ehole	Hamlet
Manganese^{54} (dis/min/kg)				
This paper	47 ± 3	82 ± 7	78 ± 4	---
Literature	---	100 ± 13[a]	---	---
Sodium^{22} (dis/min/kg)				
This paper	53 ± 6	90 ± 6	74 ± 4	78 ± 9
Literature	---	90 ± 10[a]	---	---
Aluminum^{26} (dis/min/kg)				
This paper	43 ± 3	57 ± 2	33 ± 3	52 ± 7
Literature	---	60 ± 6[a]	---	---
Scandium^{46} plus Cobalt$^{56 + 58}$ (dis/min/kg)				
This paper	11 ± 4	---	---	---
Potassium (per cent)				
This paper	0.069 ± 0.003	0.089 ± 0.004	0.083 ± 0.003	0.096 ± 0.008
Literature	---	0.0896 ± 0.0023[b] 0.0046 0.116 ± 0.008[c]	---	---

[a]Honda, M., S. Umemoto, and J. R. Arnold, J. Geophys. Res. 66, 3541-3546 (1961).

[b]Reynolds, J. H., private communication with R. E. Folinsbee.

[c]Baadsgaard, H., F. A. Campbell, R. E. Folinsbee, and G. L. Cumming, J. Geophys. Res. 66, 3574-3577 (1961).

*Most values here are an average of the hand calculation and the IBM calculation (see Table 4).

and Mn^{54}, it was measured at about 58 days and Sc^{46} and $Co^{56 + 58}$ would be expected to contribute to the 0.8-Mev photopeak. Since these spectra are complicated by the presence of several gamma-ray lines, it was considered desirable to process the Bruderheim and Harleton data with an IBM Model 704 code written by Summers and Simpson (20) as a check on the hand calculations. This code makes a least square best fit between the experimental spectrum of the meteorite and a library of reference spectra (i.e., mockups made with each of the activities present). Table 4 is a comparison of these two measurements on Bruderheim and Harleton. Results of the calculation of K^{40}, Al^{26}, Mn^{54}, Na^{22}, Sc^{46}, and $Co^{56 + 58}$ by these two techniques agreed within the estimated error of a few per cent with the exception of the Na^{22} in Harleton, where the IBM code value was about 17 per cent higher than the hand calculation. No real explanation is known for this discrepancy although, when testing the code, fairly large errors were noted when the peaks of the mockup spectra did not fall in the same channels as the peaks of the unknown spectrum. For this reason, the machine value was disregarded and the estimated uncertainty was doubled (Table 3).

The thorium contents of the 5 achondrites showing this activity are shown in Table 5, along with literature values and values on National Bureau of Standards rock samples and tektites for comparison.

TABLE 4. COMPARISON OF IBM 704 CODE* AND HAND CALCULATION OF BRUDERHEIM AND HARLETON DATA

	Bruderheim		Harleton	
	IBM 704 Code	Hand Calculation	IBM 704 Code	Hand Calculation
$Manganese^{54}$ (dis/min/kg)	87	82	47.6	46.8
$Sodium^{22}$ (dis/min/kg)	92	90	61.9	53.0
$Aluminum^{26}$ (dis/min/kg)	58	57	43.6	42.1
Potassium (per cent)	0.090	0.089	0.0697	0.0672
$Cobalt^{56 + 58}$ and $Scandium^{46}$ (dis/min/kg)	--	--	11.0	11.9

*Taken from Summers, D., and M. Simpson, unpublished data, Kirtland Air Force Base, Albuquerque, N. M. (1960).

TABLE 5. THORIUM CONTENTS OF ACHONDRITES, TEKTITES, AND TERRESTRIAL ROCKS

Sample	Thorium Content (ppm) This Paper	Thorium Content (ppm) Literature
ACHONDRITES		
Juvinas	0.60 ± 0.04	---
Nuevo Laredo	0.47 ± 0.22	0.476[a]
Pasamonte	0.52 ± 0.04	---
Sioux County	0.35 ± 0.04	---
Stannern	0.50 ± 0.04	---
TEKTITES AND GLASSES		
Bediasites (sample I)	8.6 ± 0.4	---
Bediasites (sample II)	9.4 ± 0.4	---
Australites	12.2 ± 0.4	---
Philippinites		
Santiago	13.9 ± 0.4	---
Coco Grove	14.4 ± 0.4	---
Santa Mesa	15.1 ± 0.4	---
Pugad Babuy	15.4 ± 0.4	---
Indochinites	11.9 ± 0.5	---
Libyan Desert Glass	3.3 ± 0.2	---
TERRESTRIAL ROCKS		
NBS Triassic Diabase	2.4 ± 0.2	---
NBS Columbia River Basalt	3.9 ± 0.2	---
NBS Milford Granite	9.5 ± 0.3	9.0[b]
NBS Chelmsford Granite	23.6 ± 0.6	---
NBS Graniteville Granite	43.2 ± 0.9	---
Powdered Massachusetts Diorite	9.4 ± 0.4	---
Syenite Slab	38 ± 3	---
Biotite Granite	68 ± 4	---
New Jersey Diorite	4.5 ± 0.3	---

[a]Bate, G. L., J. R. Huizenga, and H. A. Potratz, Geochim. Cosmochim. Acta 16, 88-100 (1959).

[b]Hurley, P. M., Bull. Geol. Soc. Am. 67, 405-412 (1956).

3.3 Discussion

The constancy of the Al^{26} contents of chondrites is of interest. With few exceptions, the Al^{26} content overlaps with 50 $\pm$ 10 dis/min/kg. This suggests that the cosmic-ray exposure ages of all stone meteorites measured are long compared to the half-life of Al^{26} (about 10^6 years) and that the differences in size of the meteorites measured caused little effect in Al^{26} content, an obvious exception being the Richardton chondrite, where a factor of 2 was seen in the Al^{26} measurements of different samples. This effect could be noted if Richardton were a fairly large meteorite while in space, so that the two samples measured could have been separated, and so that the sample with the low Al^{26} content could have had considerably more shielding than the other sample.

3.3.1 Peculiarities of the Beardsley Chondrite

A number of meteorite spectra merit special discussion, the first being the Beardsley samples (Figs. 8, 9, and 10). Beardsley has been recognized as a rather strange meteorite for some time now (21). Beardsley I refers to a sample obtained from the Smithsonian Institution which was collected by Nininger 2 years after infall, as was Beardsley III which was borrowed from the Arizona State University. Beardsley II,

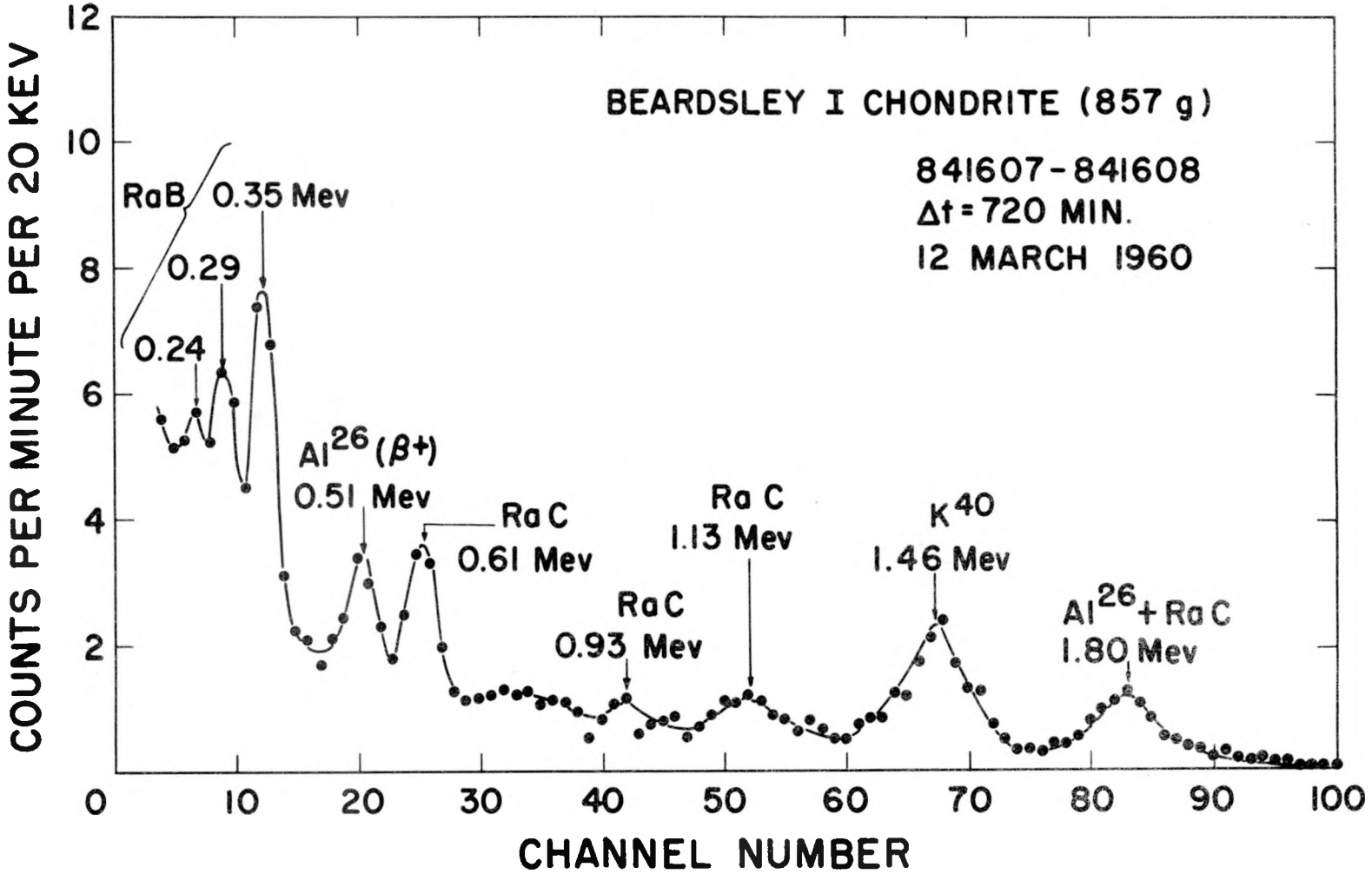

Fig. 8. Beardsley I (Smithsonian Institution) gamma-ray spectrum.

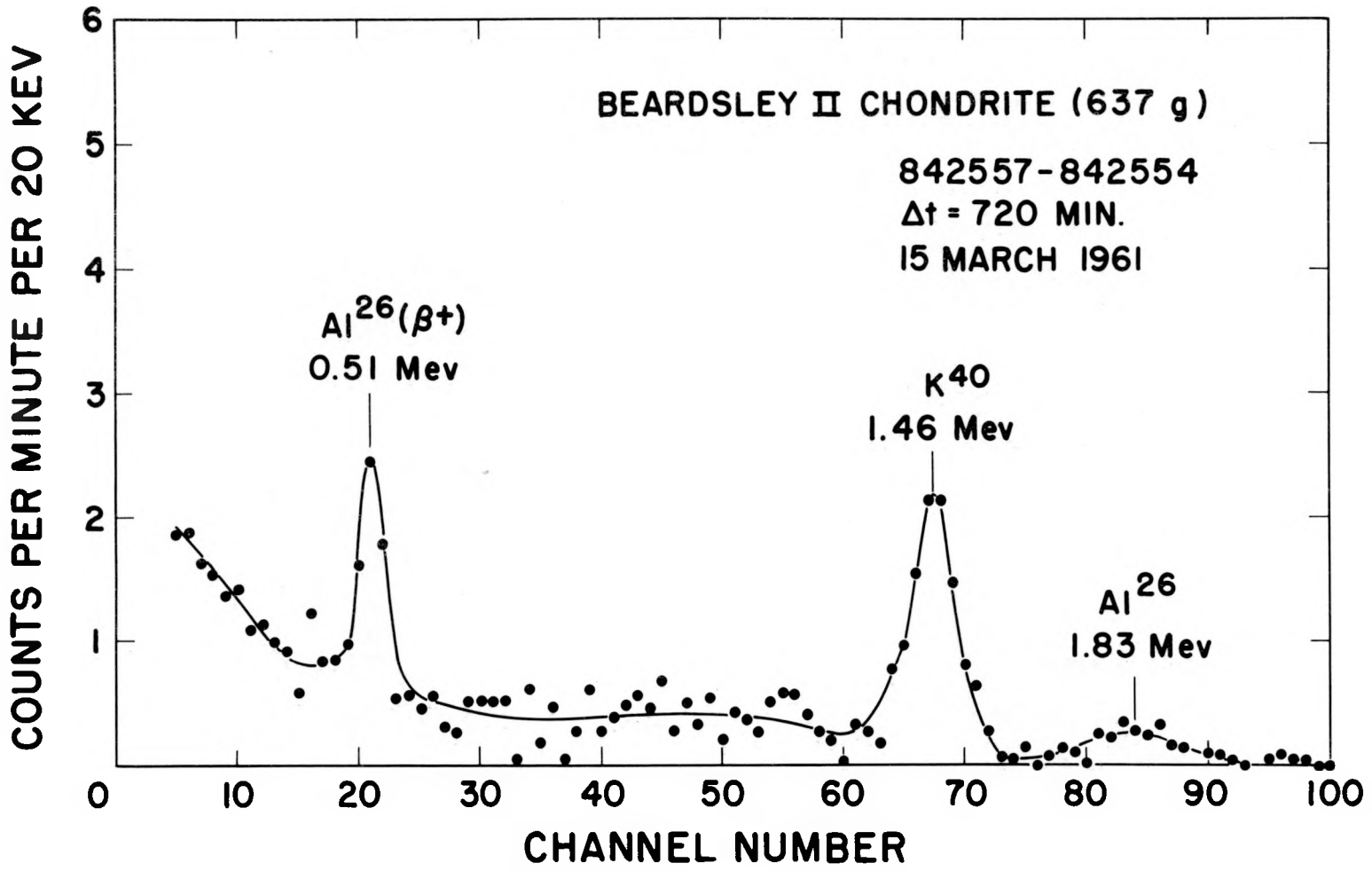

Fig. 9. Beardsley II (Michigan State University) gamma-ray spectrum.

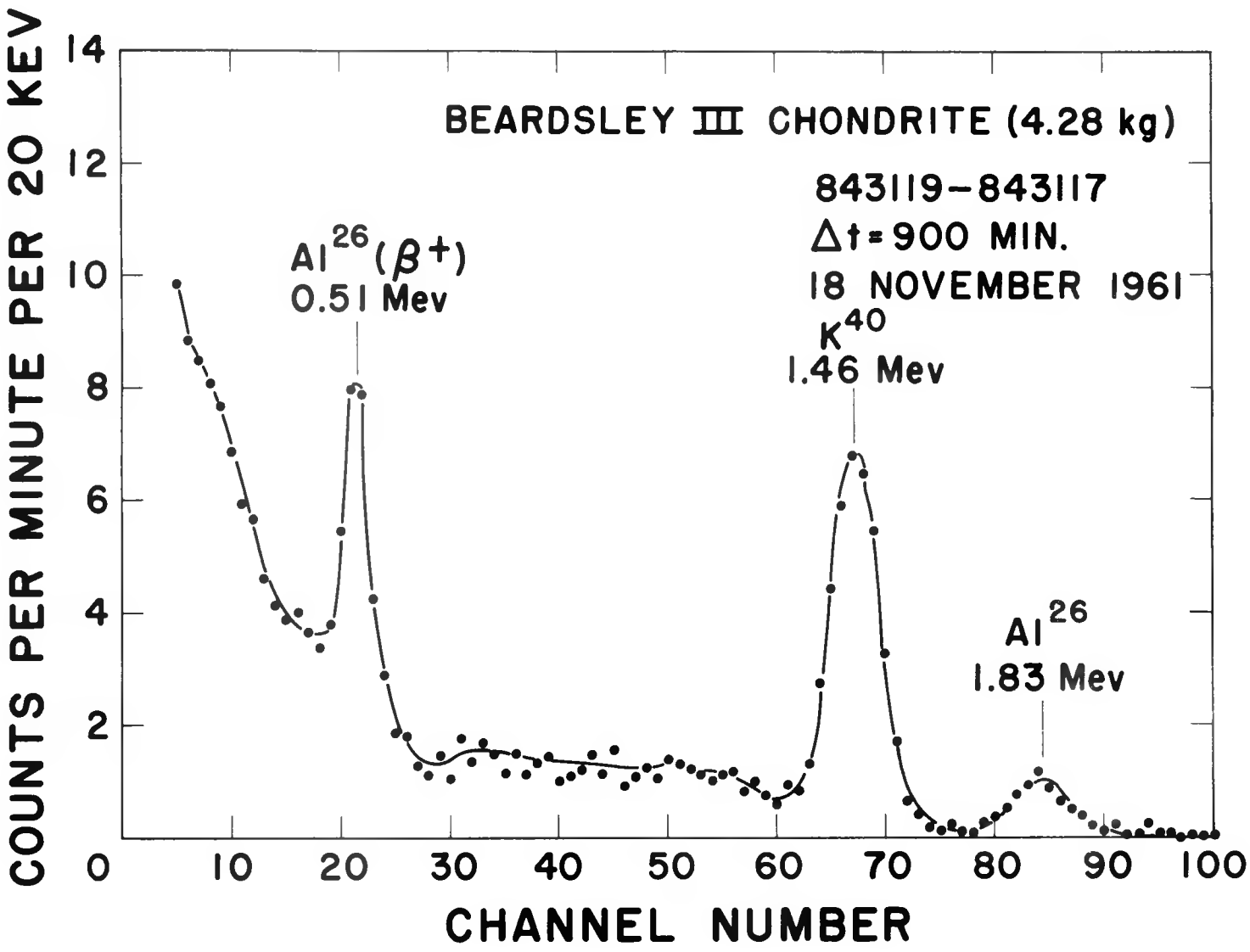

Fig. 10. Beardsley III (Arizona State University) gamma-ray spectrum.

however, was obtained from the Michigan State University and was collected the day after infall. Beardsley I showed clearly the gamma-ray spectrum of 1600-year radium and its daughter products. (Whether the radium was in equilibrium with its long-lived precursors in the uranium series could not be determined with the apparatus used.) No other chondrites measured had uranium and thorium levels that were high enough to be seen by this technique, and this is in line with the uranium/thorium analyses of others (22,23). Neither of the other 2 samples (Beardsley II and Beardsley III) showed any evidence of radium. This suggests the possibility of slight radium contamination of that particular sample of Beardsley. Extreme precautions have been used in handling all meteorites at this Laboratory so that contamination here is improbable.

Beardsley II, while free of any radium spectrum, was an unusual chondrite in other respects. Gast (21) reported a considerable difference in the potassium contents of the Michigan sample and a sample from the Nininger collection, which has been verified by our measurements. The potassium content found for Beardsley II is the highest reported for any chondrite, and it has the highest rubidium content of any meteorite, as well as an unusually high cesium content.

Beardsley III agreed in potassium content with that

reported by other workers (24,25). Its Al^{26} content was somewhat lower than Beardsley II, although it may be identical with Beardsley I since, in that case, the assay of Al^{26} was complicated by the presence of radium. All things considered, this appeared to be a rather odd meteorite.

3.3.2 Recent Falls

Bruderheim was the first stone measured here which showed cosmic-ray-induced activities other than Al^{26}; Na^{22} and Mn^{54} were both identified and assayed (26). The agreement between results on Bruderheim obtained at this Laboratory on an intact sample and results obtained at the University of California at La Jolla by Honda, Umemoto, and Arnold (27), using radiochemical separation, is very good. Subsequently, Harleton, Ehole, and Hamlet showed these activities. Harleton also showed some evidence of $Co^{56 + 58}$ and Sc^{46} in the 0.8-Mev gamma-ray photopeak. This was suggested in the decay of the 0.8-Mev photopeak. The activity was calculated by using the following simultaneous equations at two different times (t = 30 days, and t = 72 days):

$$e^{-\lambda t_1} (Co^{56 + 58} + Sc^{46}) + e^{-\lambda t_1} (Mn^{54}) = \text{c/min in 0.8-Mev photopeak at } t_1$$

$$e^{-\lambda t_2} (Co^{56 + 58} + Sc^{46}) + e^{-\lambda t_2} (Mn^{54}) = \text{c/min in 0.8-Mev photopeak at } t_2$$

Solution of these equations gives the activities of the Mn^{54} and $Co^{56 + 58}$ and Sc^{46} present. The $Co^{56 + 58}$ and Sc^{46} were assumed to have a half-life of 80 days. A Harleton spectrum is shown in Fig. 11 as representative of this group. Note that only the predominant activities are labeled. The "peak" at 0.73 Mev is thought to be the addition of the back-scatter peak and the 0.51-Mev annihilation photon (17).

3.3.3 Comparison of "Exact" and "Approximate" Mockups

Both "exact" and "approximate" mockups were used in several cases. In the La Lande chondrite, where agreement between the two results was good, the approximate mockup was made with the meteorite on hand. A determination with an approximate mockup by Van Dilla was compared with the result obtained in this report by using an exact Rezolin mockup.

An approximate mockup of Sylacauga was made by examining pictures of the sample from several views with a ruler in the picture. Agreement, in this case, was poor (the approximate potassium content = 0.083 per cent as opposed to 0.069 per cent using the exact mockup) and was undoubtedly due to the fact that the approximate mockup did not actually

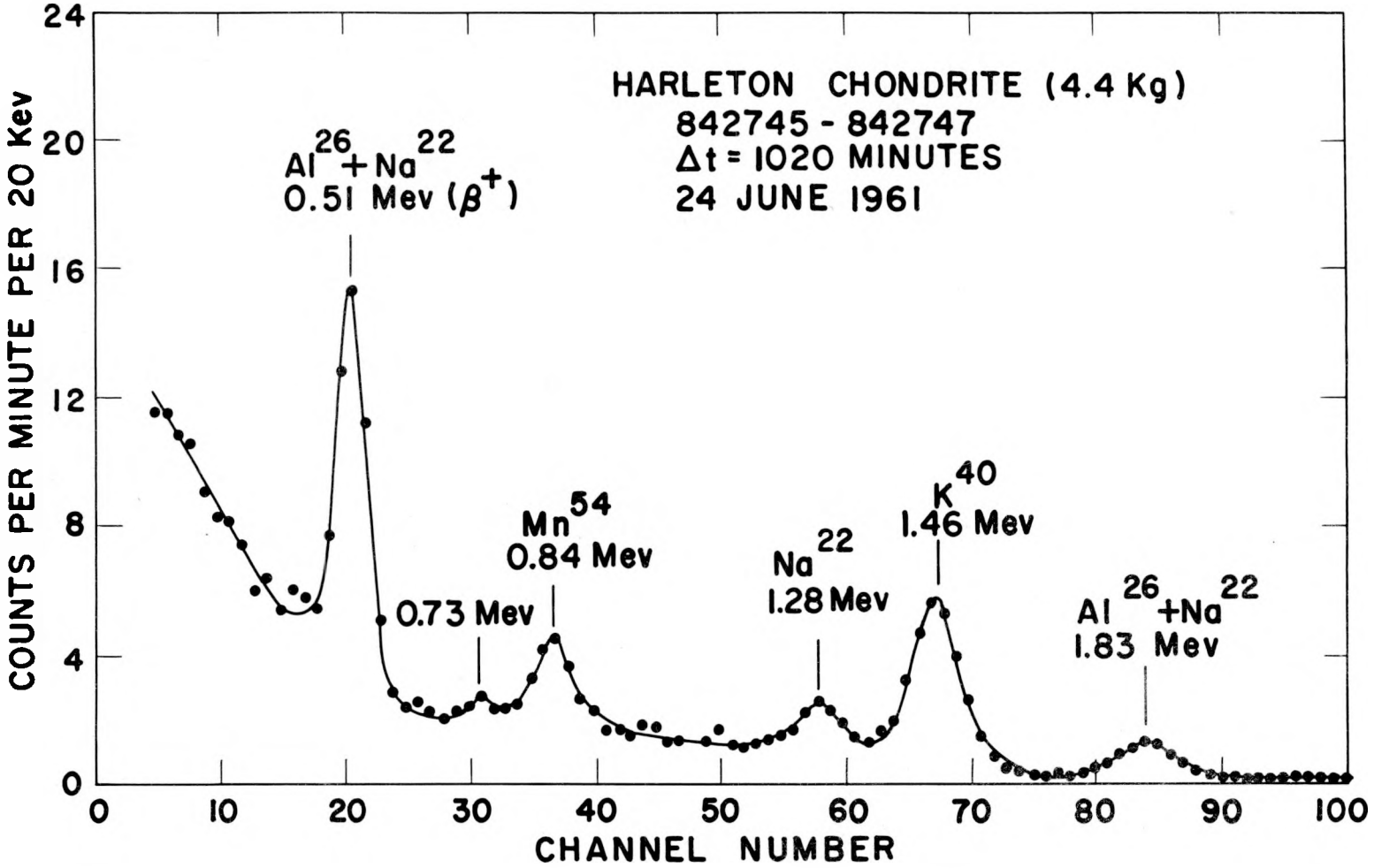

Fig. 11. Harleton chondrite gamma-ray spectrum.

coincide very closely to the geometry of the meteorite. An approximate determination on the Cavour chondrite was made by comparing it with several other meteorites of roughly the same size and known potassium and Al^{26} contents. This was compared with values obtained by Van Dilla with an approximate mockup, again made with the meteorite on hand. Agreement between the two was good; both were in agreement with the potassium content found by Maynes.

3.3.4 Carbonaceous Chondrites

Four carbonaceous chondrites were measured. Agreement of potassium content was good in all cases, except with the Murray sample, where the LASL value was about 50 per cent higher than the two values of other workers. No explanation for this discrepancy is known.

3.3.5 Achondrites

The achondrites yielded the only detectable thorium (see Fig. 12, which shows the Stannern spectrum for example), as well as the highest and lowest potassium contents found in this study. Agreement between the potassium contents for achondrites found in this report and at other laboratories was fairly good. Edwards (28) reported that the potassium content varied considerably in Bishopville, and this explains

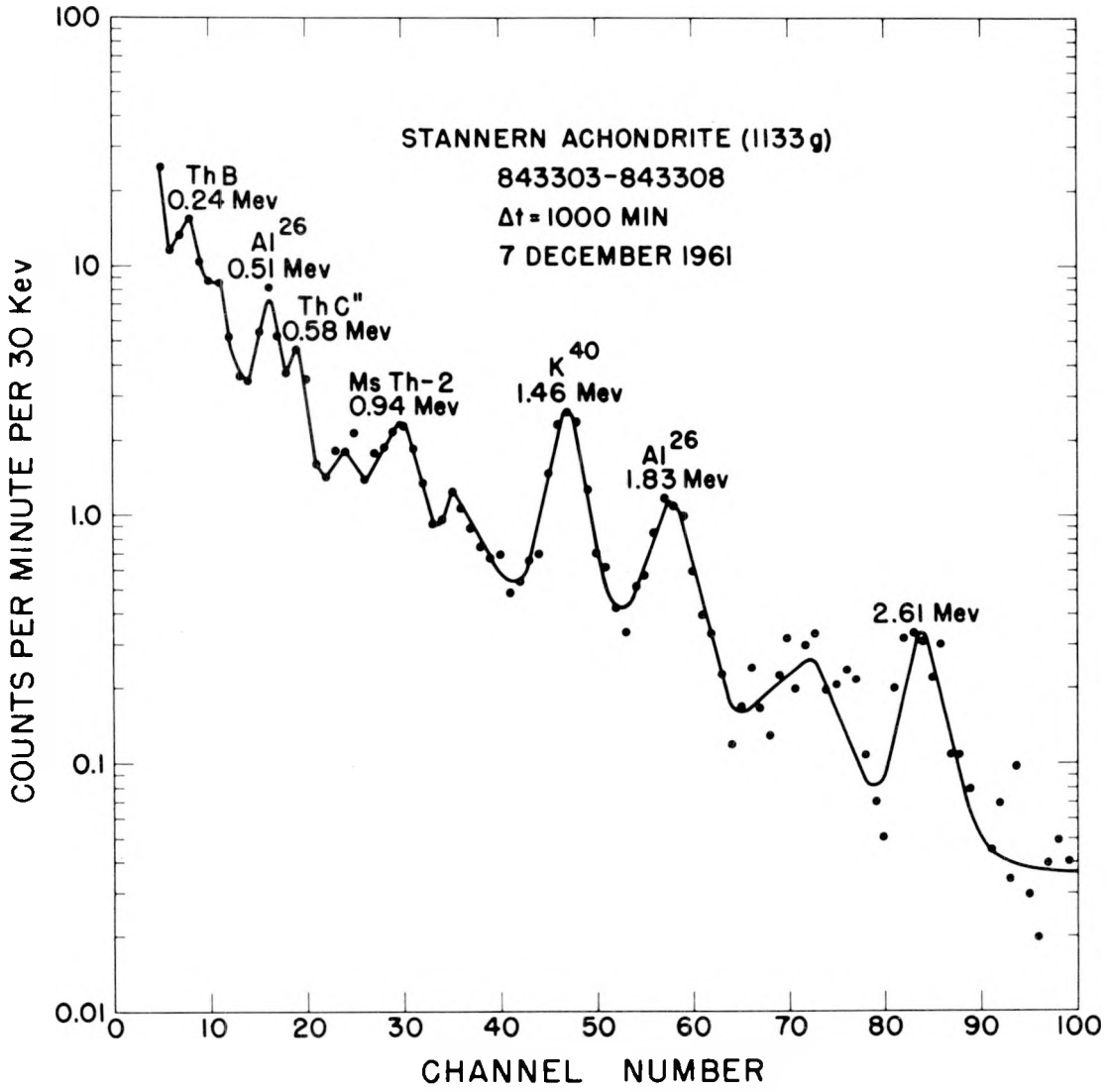

Fig. 12. Stannern achondrite gamma-ray spectrum.

any discrepancy in that meteorite. The others have relatively small amounts of potassium and are, therefore, harder to determine with comparable accuracy to chondrites. The value for the potassium content of Nuevo Laredo reported herein was rather uncertain; however, within the estimated error, it agreed with the literature values. The amount of this meteorite available for study was only about one-tenth to one-fifth of the desired amount, thus accounting for the large uncertainty in the measurement. Five of the 9 achondrites measured showed definite thorium and some evidence of uranium. The comparison of the values obtained here and those found in the literature, while rare, showed fair agreement.

3.3.6 Comparison of the Potassium Contents of Chondrites

Edwards and Urey (24) measured 21 chondrites and found an average potassium content of 0.085 per cent, with all observed values between 0.058 and 0.102 per cent. Urey (11) reported that his values were changed as a result of a correction pointed out by Geiss. His corrected value for the potassium content was 0.081 per cent. This paper reports measurements on 25 chondrites, and it has been found that the average potassium content was 0.081 per cent, with a range of from 0.060 to 0.132 per cent. If one omits Beardsley II,

because it has been shown above to be atypical in several respects, and also St. Chinian because of small sample size and resultant uncertainty, the range then becomes 0.060 to 0.105 per cent, which confirms the findings of Edwards and Urey (24). To get a better idea of the variation of the potassium contents that were measured, these values (omitting Beardsley II and St. Chinian; Beardsley I and III and Richardton I and II plotted separately) are shown in Fig. 13 as plotted on probability graph paper, which has the property of straightening out a normal distribution curve. The median at 50 per cent is seen to be 0.081 per cent potassium, with a standard deviation of $\pm$ 0.010 per cent potassium.

In order to determine if this variation was real, rather than just an analytical error, the results were compared with those in the literature, where available. Nineteen samples of 17 meteorites were treated in this manner. The LASL values were plotted against the literature values (Fig. 14), and a least squares best fit line was drawn through the points. The data thus plotted are listed in Table 6. The estimated precision on each measurement was thought to be about $\pm$ 0.003 to 0.004 per cent potassium. The least squares best fit line intersected at y = 0.004 per cent, and most of the points are seen to lie within 0.004 per cent, which lends confidence to the estimation of error in the measurements.

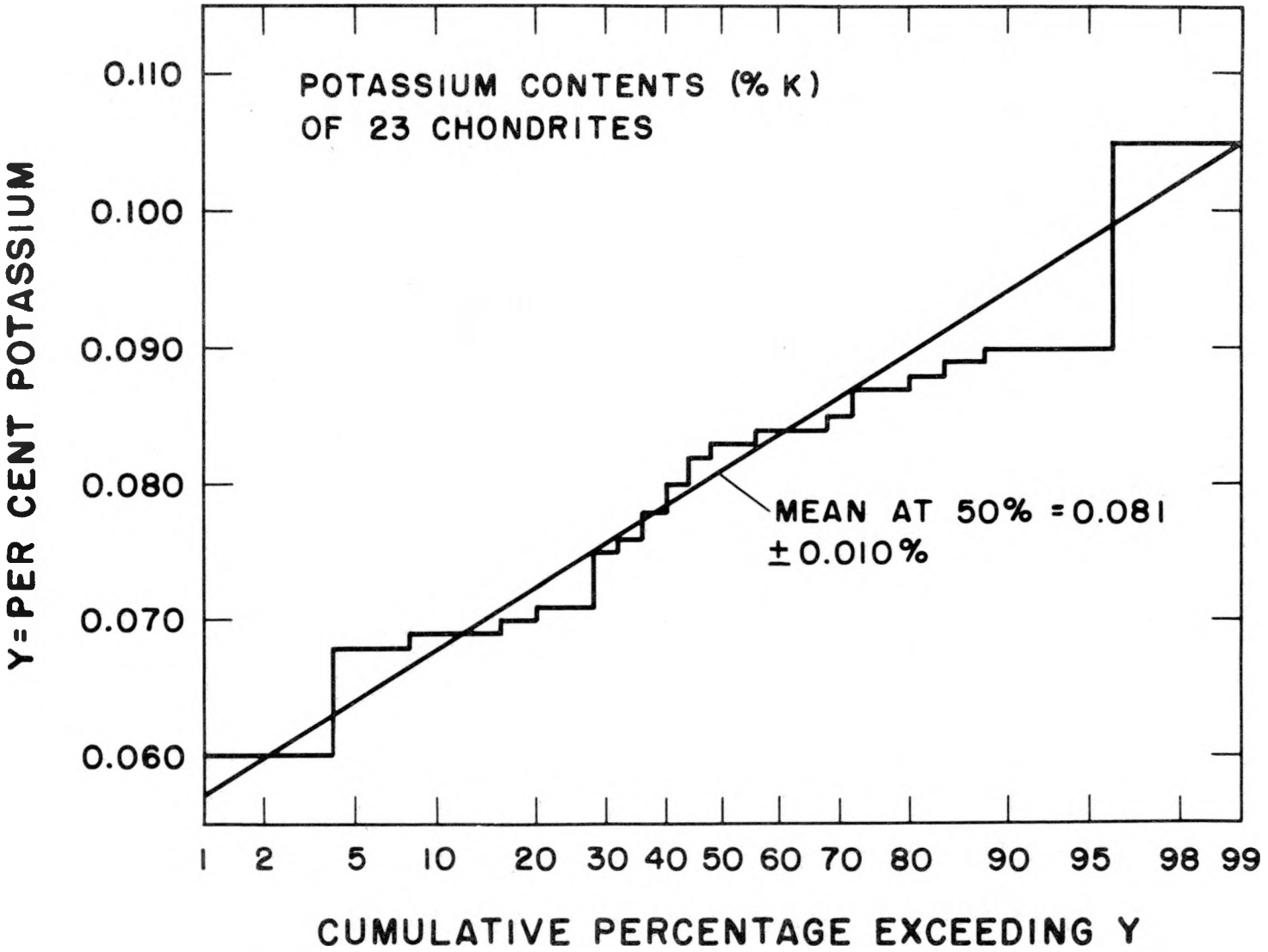

Fig. 13. Variation of the potassium contents of chondrites.

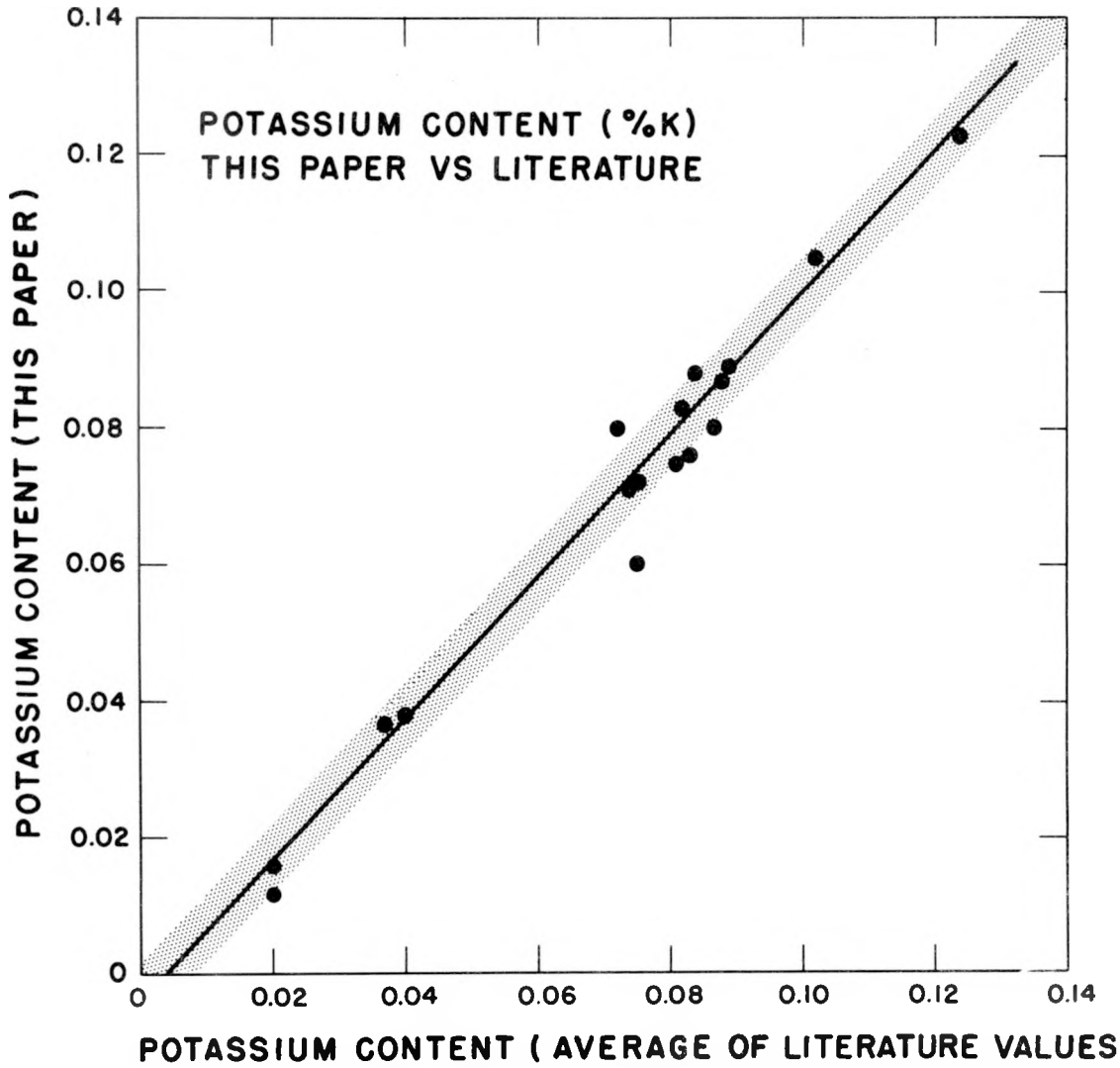

Fig. 14. Comparison of the potassium contents of stone meteorites (values in this report versus literature values).

TABLE 6. COMPARISON OF LASL VALUES WITH LITERATURE VALUES

Meteorite	Per Cent Potassium This Paper	Per Cent Potassium Others (average)	Other Values Minus LASL Values
Beardsley I	0.090	0.0906	+ 0.0006
Beardsley II	0.125	0.124	+ 0.001
Beardsley III	0.105	0.102	- 0.003
Bruderheim	0.089	0.102	+ 0.013
Cavour	0.071	0.075	+ 0.003
Forest City	0.075	0.081	+ 0.006
Holbrook	0.083	0.082	- 0.001
Ladder Creek	0.087	0.091	+ 0.004
La Lande	0.060	0.075	+ 0.015
Mocs	0.090	0.087	- 0.003
Modoc	0.088	0.084	- 0.004
Ness	0.080	0.072	- 0.008
Potter	0.071	0.074	+ 0.003
Richardton	0.076	0.083	+ 0.007
Felix	0.037	0.037	0
Indarch	0.087	0.088	+ 0.001
Mighei	0.038	0.040	+ 0.002
Moore County	0.015	0.020	+ 0.005
Norton County	0.012	0.020	+ 0.008

The formula of the least squares best fit line is:

$$K_{(LASL)} = 1.026\ K_{(Others)} - 0.004$$

If the potassium content of chondrites is a normal distribution, and if the individual values listed in this report have a random error of about $\pm$ 5 per cent, the average of the 23 should be good to about 1 per cent. This is also true of Edwards and Urey's average (24). The agreement is in support of both methods.

Since the variation of the potassium contents of chondrites, as noted in Fig. 13, was 12 per cent, and since the analytical precision was about 5 per cent, the actual variation was 11 per cent (the square root of the difference in the squares of the standard deviations).

3.3.7 Measurement of National Bureau of Standards Rock Samples

As a further check on the method for measuring the potassium content of stone meteorites on the crystal spectrometer, 7 NBS rock standards were analyzed for potassium and compared with the NBS results. Valle Grande obsidian (of local origin) was also measured and compared with a value found in the literature (29). The NBS rock samples were in powdered form and were counted in standard pint-sized

plastic containers. Mockups for these samples were made by mixing a known amount of KCl with powdered dunite so that the weight of the mockup equaled the weight of the sample. In all cases, the geometry of the sample and mockup was similar. All samples and mockups were counted on the 7-1/2 x 4-in. NaI (Tl) crystal spectrometer for counting times of 30 minutes to 3 hours. Quantitative comparisons of the K^{40} photopeaks of the mockups and samples were made. The radium and thorium contributions to the K^{40} photopeak were removed by subtracting the area enclosed by a straight line drawn beneath the photopeak (Fig. 15). The results of these measurements are shown in Table 7, and it can be concluded that errors existing in the method are small. Terrestrial rocks can be assayed with good accuracy with rather short counting times.

3.3.8 Comparison of Aluminum^{26} in Stone Meteorites

There is a fairly striking constancy in the Al^{26} contents of the 24 chondrites, 4 carbonaceous chondrites, and 9 achondrites reported, especially the chondrites, the Al^{26} dis/min/kg being almost invariably 50 $\pm$ 10.

The production mechanism for Al^{26} in stone meteorites is probably the spallation reaction on silicon and aluminum. It was, therefore, thought desirable to examine the Al^{26}

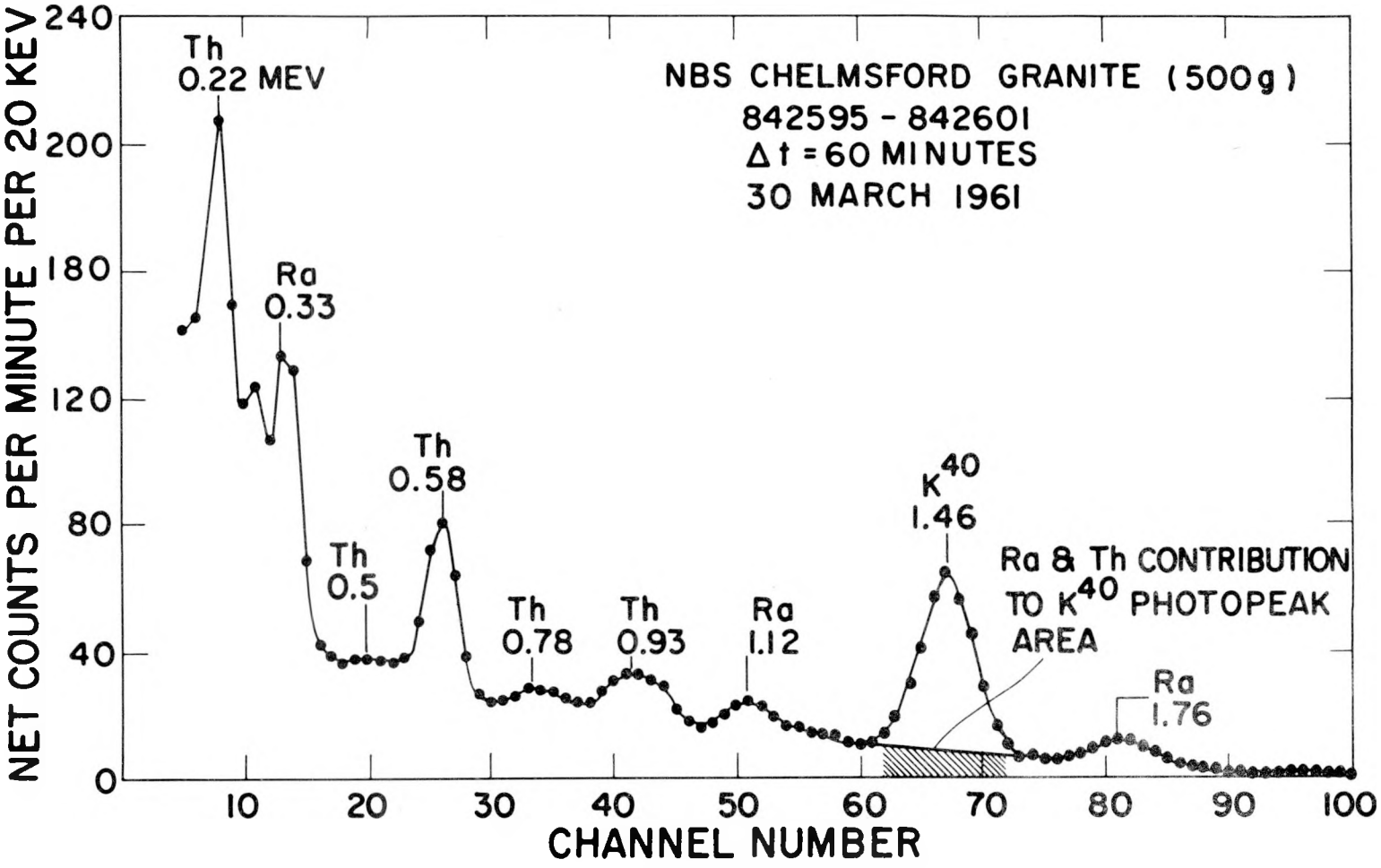

Fig. 15. Gamma-ray spectrum of NBS Chelmsford granite.

TABLE 7. POTASSIUM CONTENT OF TERRESTRIAL ROCKS

Sample	Per Cent Potassium, LASL	Per Cent Potassium, Others
NBS Milford Granite	3.33	3.32
NBS Triassic Diabase	0.46	0.48
NBS Columbia River Basalt	0.83	0.81
NBS Chelmsford Granite	4.69	4.60
NBS Graniteville Granite	3.82	3.81
NBS Gabbro Diorite	1.35	1.30
NBS Dunite	None detected	0.00
**********	**********	**********
Valle Grande Obsidian	3.35	3.56*

*Taken from Faul, H., ed., Nuclear Geology, John Wiley and Sons, Inc., New York, N. Y. (1954), p. 91.

to silicon-plus-aluminum ratio in these stone meteorites. This is the discussion which follows. The results of the measurements are shown in Table 8. The silicon and aluminum analyses were not run at this Laboratory and were either taken from the open literature or from people who were kind enough to furnish their data prior to publication. The reference column in Table 8 refers to the silicon and aluminum analyses. Whereas Table 8 includes all of the Al^{26} measurements that were made (except St. Chinian), silicon and aluminum analyses were not always available. Table 9 lists all the measurements for which silicon and aluminum analyses were available, except for those cases where thorium was detected. The analyses which showed thorium were not used because of the possibility of uranium in the Al^{26} 1.83-Mev photopeak. Figure 16 shows on probability paper a plot of the Al^{26} dis/min/kg to silicon-plus-aluminum ratio for the 24 stones for which analyses were available. It is seen from this graph that the mean at 50 per cent is 245, with a standard deviation of $\pm$ 42. An arithmetic average of these same data gives 242 for the Al^{26} to silicon-plus-aluminum ratio. The standard deviation of a finite series of observations is:

$$\sigma = \sqrt{\frac{\Sigma \delta_i^2}{n - 1}} = \sqrt{\frac{51{,}613}{26}} = 44.5$$

TABLE 8. SILICON, ALUMINUM, AND ALUMINUM^{26} CONTENTS OF STONE METEORITES

Meteorite	Silicon (per cent)	Aluminum (per cent)	Silicon plus Aluminum (per cent)	Aluminum^{26} (dis/min/kg)	Aluminum^{26} (dis/min/kg) Silicon plus Aluminum	Reference
CHONDRITES						
Abee	18.46	1.13	19.59	51	260	a
Achilles	--	--	20.00	50	250	b
Admire Stone	--	--	--	--	220	b
Archie				42		
Beardsley I	17.10	1.03	18.13	50	276	c
Beardsley II	17.10	1.03	18.13	55	304	c
Beardsley III	17.10	1.03	18.13	39	215	c
Bruderheim	18.65	0.99	19.71	57	289	d
	18.61	1.16	--	--	---	e
	--	--	--	60	300	b
Calliham				53		
Cavour	16.62	1.06	17.68	46	260	c
Cherokee Springs				40		
Ehole				33		
Forest City	17.40	1.17	18.57	35	189	c
Hamlet	--	--	--	52	---	
Harleton	18.61	1.13	19.74	43	218	f
Holbrook	18.74	0.74	19.48	58	298	g
Ladder Creek	17.98	1.10	19.08	34	178	c

TABLE 8 (continued)

Meteorite	Silicon (per cent)	Aluminum (per cent)	Silicon plus Aluminum (per cent)	$Aluminum^{26}$ (dis/min/kg)	$Aluminum^{26}$ (dis/min/kg) Silicon plus Aluminum	Reference
La Lande	18.09	1.19	19.28	49	254	c
Mocs				52		
Modoc	18.63	1.18	19.81	52	262	c
Morland	15.51	3.28	18.79	47	250	h
Ness				56		
Pantar	17.62	1.13	18.75	53	282	c
Plainview	16.30	1.41	17.71	56	316	i
Potter				52		
Richardton I	16.64	1.05	17.69	52	294	j
Richardton II	16.64	1.05	17.69	29	164	j
Sylacauga				49		
CARBONACEOUS CHONDRITES						
Felix	15.85	1.55	17.40	38	218	k
Indarch	16.47	0.68	17.15	40	233	k
Mighei	12.76	1.21	13.97	26	186	k
Murray	13.40	1.16	14.56	44	302	k
ACHONDRITES						
Bishopville	25.80	0.88	27.33	63	231	l
	26.55	1.43				m
Juvinas[S]	22.89	7.10	29.99	98	326	n
Moore County	22.47	5.55	28.02	55	196	o

TABLE 8 (continued)

Meteorite	Silicon (per cent)	Aluminum (per cent)	Silicon plus Aluminum (per cent)	$Aluminum^{26}$ (dis/min/kg)	$Aluminum^{26}$ (dis/min/kg) Silicon plus Aluminum	Reference
Norton County	25.40	0.32	25.72	53	206	k
Nuevo Laredo[s]				60		
Pasamonte[s]	22.51	7.40	29.91	69	230	p
Peña Blanca	26.68	0.11	26.79	48	179	q
Sioux County[s]				90		
Stannern[s]	22.39	5.84	28.23	96	329	r

NOTE: All the analyses used here, which were rejected by Urey, H. C., and H. Craig, Geochim. Cosmochim. Acta 4, 36-82 (1953), were rejected for reasons which were more or less unrelated to the silicon and aluminum analyses.

[a]Dawson, K. R., J. A. Maxwell, and D. E. Parsons, Geochim. Cosmochim. Acta 21, 127-144 (1960).

[b]Honda, M., S. Umemoto, and J. R. Arnold, J. Geophys. Res. 66, 3541-3546 (1961).

[c]Maynes, D., Private communication.

[d]Baadsgaard, H., F. A. Campbell, R. E. Folinsbee, and G. L. Cumming, J. Geophys. Res. 66, 3574-3577 (1961).

[e]Duke, M., D. Maynes, and H. Brown, J. Geophys. Res. 66, 3557-3563 (1961).

[f]Clarke, R. S., A preliminary report on the Chemical Composition of the Harleton, Texas meteorite (preprint).

[g]Mason, B., and H. B. Wiik, Geochim. Cosmochim. Acta 21, 276-283 (1961).

[h]Nininger, H., Trans. Kansas Acad. Sci. 39, 179-182 (1936); rejected by Urey, H. C., and H. Craig, Geochim. Cosmochim. Acta 4, 36-82 (1953).

[i]Merrill, G. P., Proc. U.S. Natl. Museum 52, 419-422 (1917); rejected by Urey, H. C., and H. Craig, Geochim. Cosmochim. Acta 4, 36-82 (1953).

[j]Quirka, T. T., J. Geol. 27, 431-448 (1919); rejected by Urey, H. C., and H. Craig, Geochim. Cosmochim. Acta 4, 36-82 (1953).

TABLE 8 (Footnotes continued)

[k]Wiik, H. B., Geochim. Cosmochim. Acta 9, 279-289 (1956).

[l]Rammelsberg, G., Monatsber. Deut. Akad. Wiss., Berlin, 895 (1953); cited in Urey, H. C., and H. Craig, Geochim. Cosmochim. Acta 4, 36-82 (1953).

[m]Merrill, G., Mem. Nat. Acad. Sci. 14 (First Memoir), 13 (1925); analysis by Whitfield; taken from Urey, H. C., and H. Craig, Geochim. Cosmochim. Acta 4, 36-82 (1953).

[n]LaCroix, A., Arch. Mus. Hist. Nat., Paris (Ser. 6), 45 (1952); Compt. Rend. 181, 747 (Raoult); taken from Urey, H. C., and H. Craig, Geochim. Cosmochim. Acta 4, 36-82 (1953).

[o]Hess, H. H., and E. P. Henderson, Am. Mineralogist 34, 494-507 (1949).

[p]Foshag, W., Am. J. Sci. 35, 374-382 (1938).

[q]Lonsdale, J., Am. Mineralogist 32, 354-364 (1947).

[r]Merrill, G., Mem. Nat. Acad. Sci. 14 (First Memoir), 22; analysis by Whitfield; taken from Urey, H. C., and H. Craig, Geochim. Cosmochim. Acta 4, 36-82 (1953).

[s]Not used in final analysis because of the possibility of uranium (RaC) contributing to the Al^{26} peak.

TABLE 9. DATA FROM TABLE 8

No.	Meteorite	$Aluminum^{26}$ (dis/min/kg) Silicon plus Aluminum	δ	δ^2	Meteorite Type
1	Richardton II	164	-78	6084	Chondrite
2	Ladder Creek	178	-64	4096	Chondrite
3	Peña Blanca	179	-63	3969	Achondrite
4	Mighei	186	-56	3136	Carbonaceous Chondrite
5	Forest City	189	-53	2809	Chondrite
6	Moore County	196	-46	2116	Achondrite
7	Norton County	206	-36	1296	Achondrite
8	Beardsley III	215	-27	729	Chondrite
9	Felix	218	-24	576	Carbonaceous Chondrite
10	Harleton	218	-24	576	Chondrite
11	Admire Stone	220	-22	484	Pallasite
12	Bishopville	231	-11	121	Achondrite
13	Indarch	233	- 9	81	Carbonaceous Chondrite
14	Achilles	250	8	64	Chondrite
15	Morland	250	8	64	Chondrite
16	La Lande	254	12	144	Chondrite
17	Abee	260	18	324	Chondrite
18	Cavour	260	18	324	Chondrite
19	Modoc	262	20	400	Chondrite

TABLE 9 (continued)

No.	Meteorite	$Aluminum^{26}$ (dis/min/kg) Silicon plus Aluminum	δ	δ^2	Meteorite Type
20	Beardsley I	276	34	1156	Chondrite
21	Pantar	282	40	1600	Chondrite
22	Bruderheim	294	52	2704	Chondrite
23	Richardton I	294	52	2704	Chondrite
24	Holbrook	298	56	3136	Chondrite
25	Murray County	302	60	3600	Carbonaceous Chondrite
26	Beardsley II	304	62	3844	Chondrite
27	Plainview	316	74	5476	Chondrite
		Average = 242		Σ = 51,613	

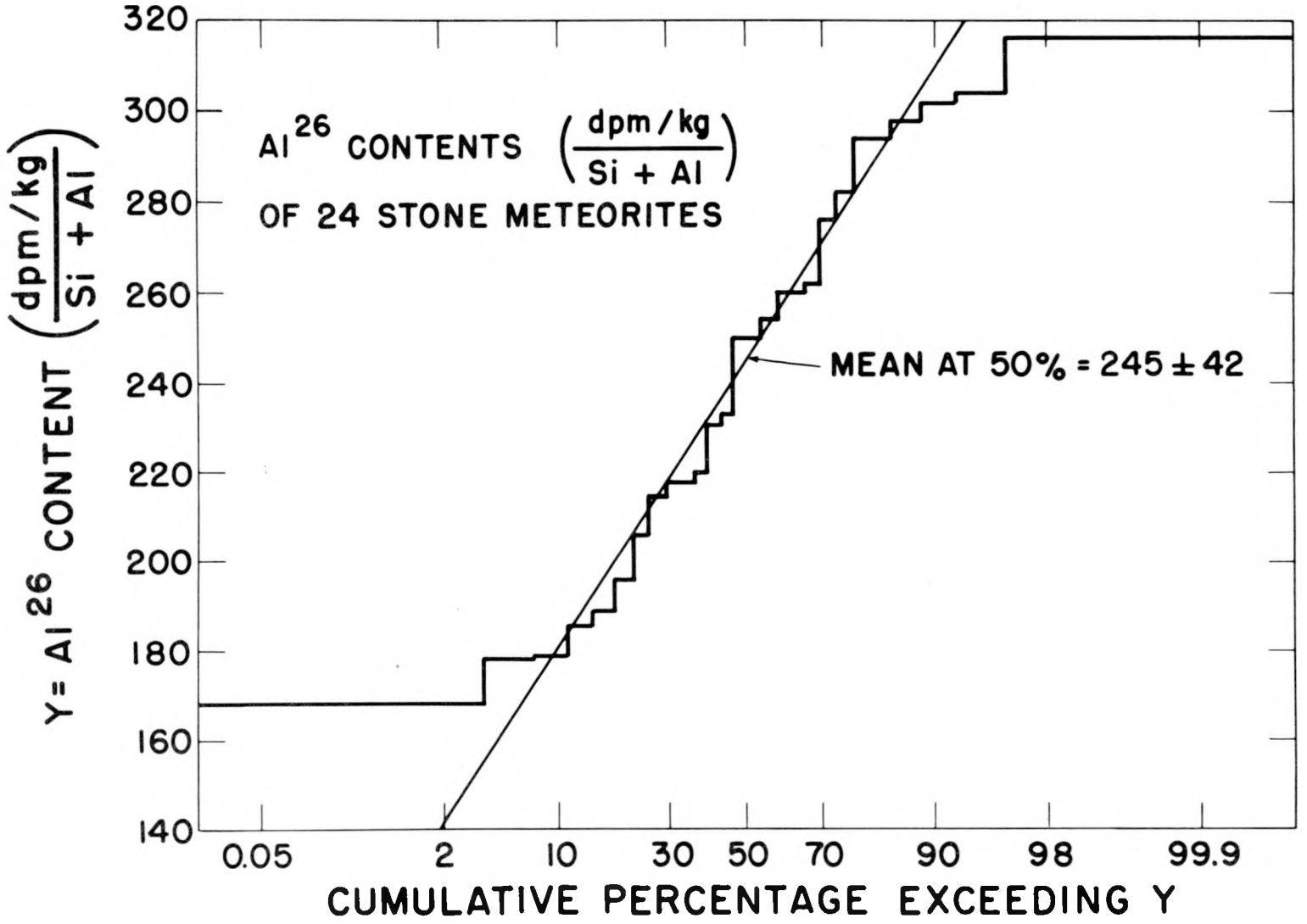

Fig. 16. Variation of the Al^{26} contents of stone meteorites.

where σ = standard deviation; δ_i = variation of the i^{th} measurement from the average; and n = number of measurements.

The two methods are seen to yield essentially the same number, about 245 $\pm$ 45. This agreement lends confidence that the number of measurements can be handled statistically. Since the estimated standard deviations of the individual measurements of Al^{26} are thought to be $\leq$ 10 per cent in all cases, the real variation of Al^{26} in stones (per silicon and aluminum) can be found. The observed standard deviation is $\pm$ 18 per cent; therefore, the minimum standard deviation due to actual variation in the samples is:

$$\sigma_m = \sqrt{18^2 - 10^2} = 15 \text{ per cent.}$$

This suggests that the actual variation in Al^{26} content in the stone meteorites measured, which may be caused by such factors as shielding effects and variation of cosmic-ray flux, etc., is about 15 per cent.

Table 10 shows the Al^{26} measurements that have been run by other investigators compared with these results. Agreement is very good on Achilles, Bruderheim, and Ladder Creek; the variations noted in Plainview and Richardton are probably real. The Al^{26} content of Ladder Creek I was calculated here by two different techniques. Agreement was fair in all cases. The thought that the standard deviation

TABLE 10. COMPARISON OF ALUMINUM^{26} CONTENTS IN STONE METEORITES

Meteorite	LASL (Al^{26} dis/min/kg)	Others (Al^{26} dis/min/kg)	Reference
Achilles	50 ± 5	50 ± 5	d
Bruderheim	57 ± 2	60 ± 6	d
Ladder Creek I	29 ± 5[a]	32 ± 3[b]	e
	33 ± 3[c]		
Ladder Creek II	36 ± 3		
Plainview	56 ± 5	54 ± 5	f
		68 ± 5	g
		38.5 ± 5	h
Richardton	52 ± 6	63 ± 4	g
	29 ± 3	65 ± 8	h

[a]Calculated by comparing the Al^{26} photopeak with the known K^{40} photopeak.

[b]Measured on a coincidence spectrometer [two 8 x 4-in. NaI (Tl) crystals] using the same Al^{26} mockup as this author.

[c]Calculated by using an "exact" Al^{26} mockup.

[d]Honda, M., S. Umemoto, and J. R. Arnold, J. Geophys. Res. 66, 3541-3546 (1961).

[e]Van Dilla, M. A., private communication (1961).

[f]Ehmann, W. D., and T. P. Kohman, Geochim. Cosmochim. Acta 14, 364-379 (1958).

[g]Anders, E., Geochim. Cosmochim. Acta 19, 53-62 (1960).

[h]Chakrabartty, M., Carnegie Institute of Technology Progress Report in Nuclear Chemistry, 1960-1961 (1961), pp. 68-73.

of the Al^{26} measurements is ≤ 10 per cent is supported by the comparison of potassium measurements on meteorites made at this Laboratory with those from other laboratories (see above).

CHAPTER 4

SIDERITES

The measurements on the irons herein reported are an extension of those previously reported by Van Dilla, Arnold, and Anderson (7). The previous measurements were chiefly qualitative; quantitative results are now reported.

A total of 6 siderites have been measured, but measurable activities were found in only 2 of these: Aroos and Sikhote-Alin. The Aroos siderite fell in Azerbaijan, U.S.S.R., on November 24, 1959. A 320-g slice was measured here in February 1960, 120 days after fall. The Sikhote-Alin siderite fell in the Sikhote-Alin Mountains north of Vladivostok on February 12, 1947. Manganese^{54} was the predominant radioactivity detected in Aroos, while Co^{60} was the only radioactivity detected in Sikhote-Alin.

4.1 Experimental Procedure

All the siderites were measured on a gamma-ray spectrometer using a 7-1/2 x 4-in. NaI crystal. The technique

used for quantitative assay of the Mn^{54} content of the 320-g slice of Aroos was as follows. A stack of 5 mild steel plates (2 x 2-3/4 x 1/8 in.), containing uniform Cs^{137} surface distribution, served as the mockup; this was counted in essentially the same geometry as the meteorite. The source of the Cs^{137} was a Nuclear-Chicago standard with an accuracy stated as $\pm$ 3 per cent. Since the Aroos sample and mockup were small compared to the NaI crystal, slight geometry differences and possible inhomogeneity of radioactive concentration had little effect. Small corrections were necessary for differences in self-absorption, photofraction, and energy resolution between the 0.66-Mev Cs^{137} gamma rays and the 0.84-Mev Mn^{54} gamma rays. The activity in the 1.1-Mev photopeak (probably Sc^{46} and Co^{60}) was similarly compared with the 0.84-Mev Mn^{54} photopeak.

The same method was used for the calibration of the Co^{60} contents of the 1.14-, 1.295-, and 6.21-kg samples of Sikhote-Alin except that a standardized Co^{60} solution was available which eliminated the small corrections due to the difference in energy between the sample and mockup gamma rays. The Co^{60} solution used to calibrate all Sikhote-Alin samples was calibrated against a solution which had been 4π-counted by Balagna (30). It was also compared with a Nuclear-Chicago Co^{60} standard. The two results had a discrepancy of only

1.4 per cent. The larger size of these samples would introduce somewhat more uncertainty because of geometry and possible inhomogeneity, but these effects would still be expected to be small.

The 9.5-kg sample of Sikhote-Alin presented a different problem because of its large size and irregular shape. Our solution to this problem was as follows. A thin, hard shell was made the shape of the meteorite. The procedure used to make this shell was the same as was described earlier in this report. This shell was then given to the LASL Metallurgy Group (CMB-6), who then prepared a casting of a 90 per cent copper and 10 per cent aluminum alloy. This alloy was chosen because the combination of its electron density and its physical density made its interactions with gamma rays in the region 0.5 to 2 Mev very similar to those of iron (the LASL Metallurgy Group was not set up to handle iron). At this energy, Compton absorption and scattering are virtually the only interactions taking place in the siderite and mockup. This casting was then sawed into 1/2-in. slices by the Hughes Tool Company, Engineering Research Laboratory, Houston, Texas. The cuts were taken parallel to the NaI crystal face when the mockup was in its counting position, and 1/16 in. iron plates were added between slices of the copper-aluminum alloy to make up for the amount lost in sawing. A uniform NBS-calibrated

Co^{60} solution was then pipetted in a 2-cm grid pattern on the iron plates, forming the completed mockup.

4.2 Results

Of the 6 meteorites measured, only Aroos and Sikhote-Alin showed detectable gamma-ray activities. The spectra of these two irons are given in Figs. 17 and 18, and the data are summarized in Table 11 (positive results) and in Table 12 (negative results). The Aroos spectrum showed a prominent photopeak at 0.84 Mev with suggestions of peaks at about 1.1 and 1.3 Mev. The most likely interpretation of the 0.84-Mev peak is that it was due principally to 291-day Mn^{54} with a small contribution from 73-day Co^{56} and 71-day Co^{58}. The two higher energy peaks were more uncertain, and it is likely that only the 1.1-Mev peak was significant. This peak may have been due to 84-day Sc^{46} with some contribution from Co^{60}. The 1.3-Mev peak can be explained on the basis of a very slight shift in energy calibration between the sample and background (note the "valley" of equal negative area just to the right).

Definite Co^{60} was seen in the Sikhote-Alin samples, and the results on 3 of the samples agreed; the fourth was significantly lower.

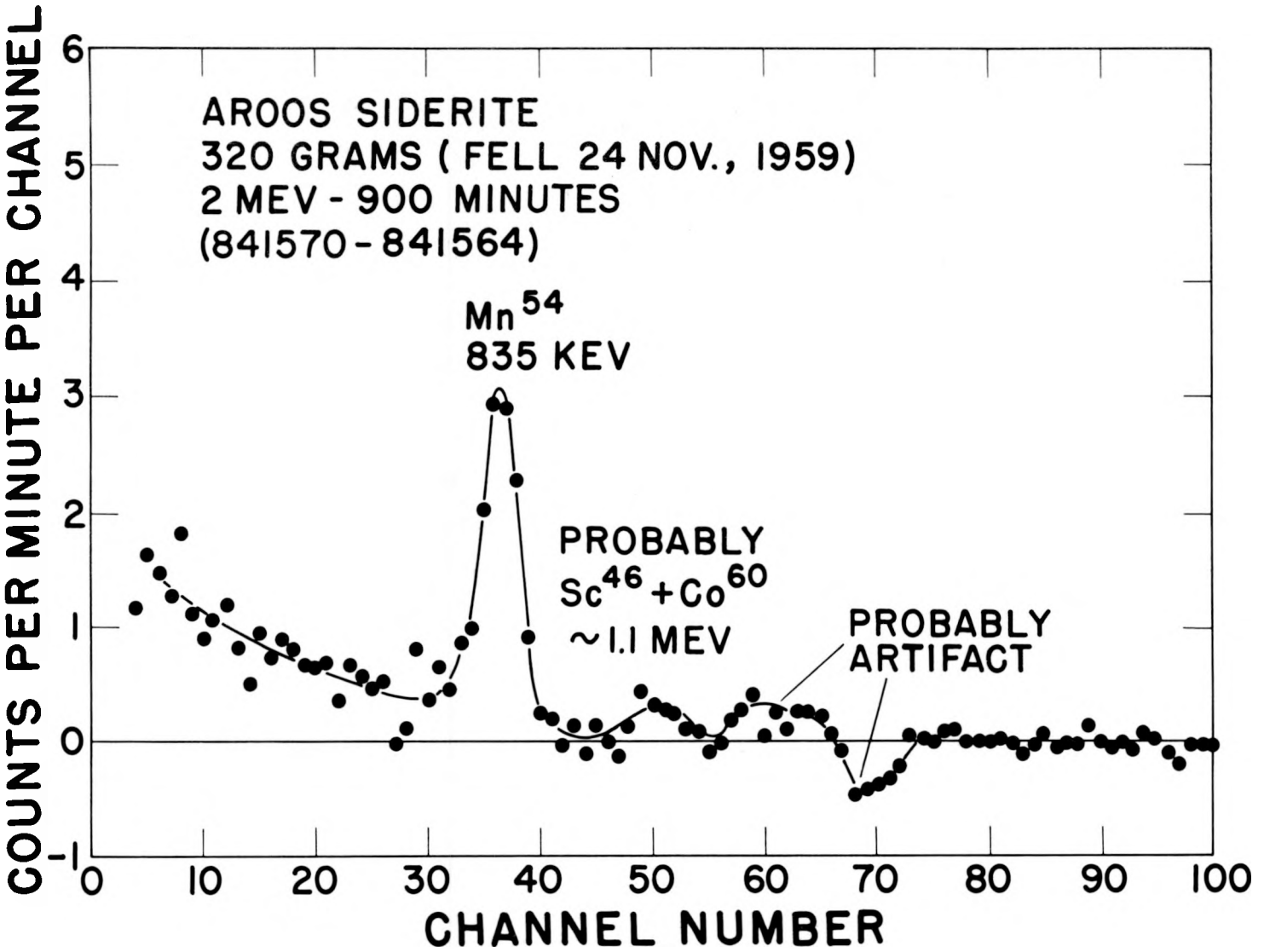

Fig. 17. Gamma-ray spectrum of the Aroos siderite.

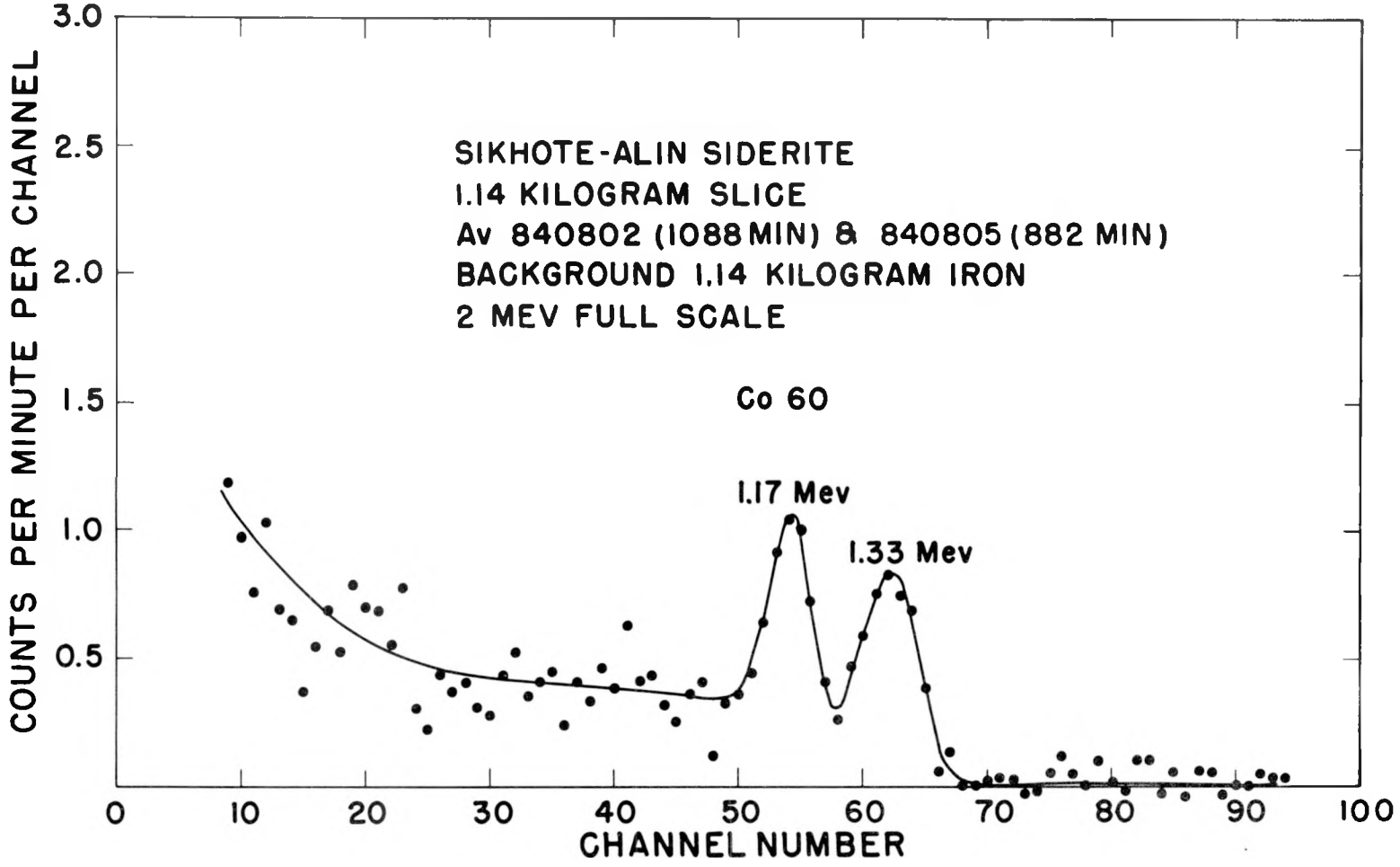

Fig. 18. Gamma-ray spectrum of the Sikhote-Alin siderite.

TABLE 11. RADIOACTIVITY IN IRON METEORITES: POSITIVE RESULTS

Meteorite	Source	Known Mass (kg)	Sample Weight (kg)	Years (Fall to Assay)	Activity Corrected to Time of Fall	
					This Paper	Other Reports
Aroos	Arnold-Krinov	150	0.320	0.33	480 ± 35 dis/min Mn^{54}/kg[a]	470 ± 47 dis/min Mn^{54}/kg[b]
					425 ± 40 dis/min Mn^{54}/kg[c]	---
					45 ± 15 dis/min Co^{60} + Sc^{46}/kg[d]	28 ± 3 dis/min Co^{60} + Sc^{46}/kg[b,d]
Sikhote-Alin	LaPaz	~100,000	1.14	12.8	382 ± 38 dis/min Co^{60}/kg	---
	LaPaz		6.21	12.8	332 ± 33 dis/min Co^{60}/kg	---
	Arnold-Krinov		1.30	13.2	207 ± 21 dis/min Co^{60}/kg	---
	LaPaz		9.50	13.6	386 ± 39 dis/min Co^{60}/kg	---

[a]Assumes only Mn^{54} present in 0.84-Mev photopeak at 120 days after fall.

[b]Honda, M., and J. R. Arnold, Geochim. Cosmochim. Acta 23, 219-232 (1961).

[c]Honda and Arnold calculation of the data presented herein, correcting for contributions of $Co^{56 + 58}$ from their data.

[d]Calculated at time of measurement (t = 120 days), rather than time of fall.

TABLE 12. RADIOACTIVITY IN IRON METEORITES: NEGATIVE RESULTS

Meteorite	Source	Sample Weight (kg)	Years (Fall to Assay)	% Co^{60} Left at Assay
Pitts	Henderson	1.13	36	0.88
Odessa	LaPaz	12.8	$\geq$ 35	$\leq$ 1.1
Cañon Diablo	Ward's N. S. E.	1.99	$\geq$ 66	$\leq$ 0.02
Toluca	LaPaz	16.6	>183	<< 0.01

4.3 Discussion

The agreement between the results presented on the Mn^{54} content of Aroos and those of Honda and Arnold (31) is very good, considering the two very different methods of measurement. If the 1.1-Mev peak is real and due to Sc^{46} and Co^{60}, then the combined activity is 45 $\pm$ 15 dis/min/kg. The data of Honda and Arnold showed that 28 $\pm$ 3 dis/min/kg of these activities was present at time of the measurement reported herein. This lends support to the interpretation of the 1.1-Mev peak. It is probable that the peak at 1.3 Mev and the negative peak at 1.5 Mev are artifacts resulting from subtraction of two spectra (sample and background) containing relatively large background K^{40} peaks recorded under slightly different energy calibration.

Manganese^{54} is probably produced mainly from the spallation reaction on iron with some contribution from nickel and cobalt. The Mn^{54} dis/min/kg iron in the Bruderheim chondrite have been shown to be very similar to the value obtained for Aroos (26,3), which is in support of this idea.

Production of Co^{60} is thought to take place by an entirely different mechanism (namely, capture by Co^{59}, 100 per cent abundant, of neutrons produced by cosmic rays and then moderated to low energies). In contrast to the Mn^{54} situation, the Co^{60} concentration in iron meteorites should be very

dependent on mass. Slowing down of neutrons in a siderite is the result of inefficient collisions with relatively heavy nuclei (mostly iron, A = 56). It is estimated that a very massive siderite (several tons in space) is necessary to produce the maximum number of low-energy neutrons. Production of Co^{60} in an iron of optimum size is expected to be about 300 dis/min/kg from theoretical considerations (7). This seems to be in fair agreement with the values herein reported.

Since the known Sikhote-Alin mass is very large ($\sim 10^5$ kg), significant variations in the Co^{60} concentration might be expected within the mass. The numbers reported here indeed bear this out. If the 2 smaller Sikhote-Alin samples loaned by Dr. LaPaz for measurement were adjacent on the original body (the cross sections of the 2 pieces were very similar) and at a quite different location from the sample obtained from Dr. Arnold, then the difference in Co^{60} concentrations could well be a mass or depth effect. These types of depth and mass effect data might yield useful information regarding the original size of the Sikhote-Alin fall. This suggests that measurement of the Co^{60} concentration in the large amount of this fall in the U.S.S.R. should be undertaken.

The difference in Co^{60} contents of Sikhote-Alin and Aroos

follows from the above discussion. Since several tons are necessary to yield the maximum number of low-energy neutrons, Aroos (150 kg recovered) would be expected to be considerably lower in Co^{60}.

Another way of expressing the data is the Co^{60} to cobalt ratio. Data available on Sikhote-Alin, Aroos, and the iron phase of Bruderheim showed the following:

Sikhote-Alin	~5 to 10 x 10^4 dis/min Co^{60}/kg cobalt
Bruderheim	1.8 x 10^4 dis/min Co^{60}/kg cobalt
Aroos	~0.4 x 10^4 dis/min Co^{60}/kg cobalt

The Aroos data were taken from Honda and Arnold (31), and the Bruderheim Co^{60} data were taken from Honda, Umemoto, and Arnold (27). The per cent cobalt in Bruderheim was that of Baadsgaard, Campbell, Folinsbee, and Cumming (32), and the per cent cobalt in Sikhote-Alin was that of Gerling and Levskii (33). Thus, the stone Bruderheim was shown to have about 4 times the concentration of Aroos; whereas, its recovered mass was about twice that of Aroos. In the Sikhote-Alin, which was large enough to have the maximum number of low-energy neutrons, it was a factor of about 4 above the Bruderheim value.

The negative results on the 4 irons shown in Table 12 are explained by the fact that the major gamma-ray emitting

isotopes (Sc^{46}, V^{48}, Cr^{41}, Mn^{54}, Co^{56+58}, Co^{57}, and Co^{60}) produced by cosmic rays in iron meteorites are short-lived (half-lives < 5.27 years) and have decayed below the limit of detection between the time of fall and time of measurement. No gamma-ray activity in the energy range 0.2 to 2 Mev was detectable in these samples with a limit of detection of about 10 gammas/min/kg.

The differences between the chondrites and the irons using the technique presented here are easily seen. Whereas the chondrites have measurable natural K^{40}, the K^{40} in irons is cosmic-ray produced and is below the limits of detection at this Laboratory. The chondrites probably have all the cosmic-ray-induced activities found in the irons, since they contain roughly 20 to 25 per cent iron by weight; in the stones, however, these activities which are produced from iron are down by a similar factor. Aluminum^{26}, which is the most prominent activity (along with K^{40}) observed in the stones by gamma-ray spectrometry, is formed by spallation of silicon and aluminum. In irons, any Al^{26} is below the limit of detection at this Laboratory. The most outstanding feature of irons older than 25 years is the complete absence of gamma emitters by this technique. For example, see Fig. 19, which is the gamma-ray spectrum of the Cañon Diablo siderite.

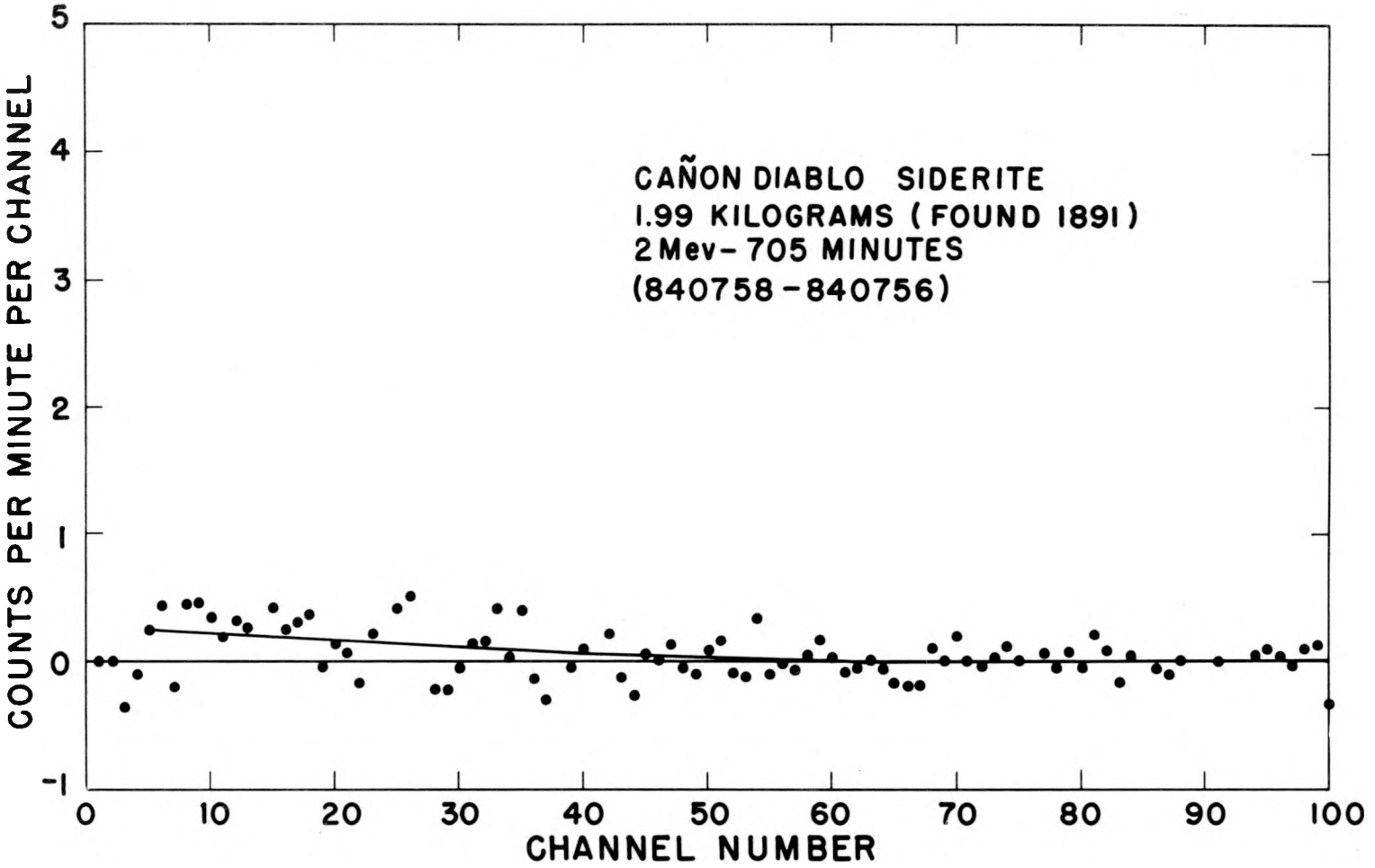

Fig. 19. Gamma-ray spectrum of the Cañon Diablo siderite.

CHAPTER 5

SUMMARY

A new, nondestructive gamma-ray spectrometry technique for the quantitative measurement of small amounts of gamma-ray emitters in meteorites has been presented.

The accuracy of the method is supported by the agreement with published values on meteorites, as well as by the determinations made on National Bureau of Standards rock samples. The accuracy obtainable by this technique is comparable with results obtained by methods of wet chemistry, but the method has the advantages that it is nondestructive and that no chemical processing is necessary. Another advantage in many applications is the fact that the method automatically averages over a fairly large sample size (~ 1 kg is ideal). The minimum size that can be handled with reasonably good accuracy is about 400 g.

The Al^{26} contents determined here are in fairly good agreement with those of other workers. The differences noted

(Richardton, for example) are probably real and are perhaps due to differences in shielding effects within the same meteorite.

The variation in potassium contents and the average value for chondrites found at this Laboratory confirm the findings of Edwards and Urey. The variation is almost certainly real.

Further problems suggested are the measurement of the variation of Al^{26} through the mass of a large stone meteorite such as the Norton County achondrite.

The uranium contents of the achondrites which contained thorium could probably be quantitated by using a coincidence spectrometer with two NaI (Tl) crystals facing each other, with the meteorite sample between the two.

The large amounts of the Sikhote-Alin siderite in the U.S.S.R. would offer a good opportunity to study the variation of Co^{60}. Especially interesting would be a study of the Co^{60} contents of samples which occupy known positions with respect to each other. Such a study might yield useful information on the neutron-capture reaction in iron meteorites. Both of these investigations might be interesting with respect to the ablation and original masses of the meteorites.

No siderolites were studied.

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Dr. Lincoln LaPaz, Director of the Institute of Meteoritics, University of New Mexico, Albuquerque, New Mexico.

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Dr. H. C. Davis, Director of the North Carolina State Museum, Raleigh, North Carolina.

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