REVIEWS OF MODERN PHYSICS VOLUME 34, NUMBER 2 APRIL, 1962

Meteorite Ages

EDWARD ANDERS

The Enrico Fermi Institute for Nuclear Studies and Departments of Chemistry and Geophysical Sciences
The University of Chicago, Chicago, Illinois, and Theoretical Division, Goddard Space Flight Center
National Aeronautics and Space Administration, Greenbelt, Maryland

CONTENTS

THERE are five principal events in the history of meteorites that can be dated by radioactivity. Stated in somewhat oversimplified terms, they are

1. Nucleosynthesis
2. Melting of meteorite parent bodies
3. Cooling of meteorite parent bodies
4. Breakup of meteorite parent bodies
5. Fall of meteorite.

The first of these is dated by extinct radioactivity, e.g., 16.4 million years I^{129}; the second and third by long-lived radioactivity, such as 4.51×10^9 yr U^{238} and 1.3×10^9 yr K^{40}; and the fourth and fifth, by cosmic-ray-induced radioactivity, such as 12.3-yr H^3, 3.08×10^5-yr Cl^{36}, and 325-yr Ar^{39}. These dating methods are discussed here in approximately the reverse order.

1. TERRESTRIAL AGES

From the time the meteorite is reduced to a fragment of meter size or smaller to the time of its fall, it is strongly irradiated by cosmic rays. These induce various nuclear reactions in the meteorite. Typically, an iron nucleus is struck by a high-energy proton, losing several light particles (protons, neutrons, and helium nuclei) in the process. The residual nucleus can be either stable, e.g., Sc^{45}, or radioactive, e.g., Cl^{36}:

$$Fe^{56}+H^1 \rightarrow Cl^{36}+H^3+2\,He^4+He^3+3\,H^1+4n. \quad (1)$$

Each residual nucleus can be reached by a variety of paths. From the amounts and proportions of these nuclides, it is possible to infer many details of the meteorite's history, for example, the time of its fall, or "terrestrial age."

The production rate H_s of a cosmic-ray-produced nuclide S, in atoms/sec g (Wänke and Vilcsek 1959; Eberhardt and Geiss 1960a),[1] is given by

$$H_s=4\pi I_0 P_s N, \quad (2)$$

where I_0 is the primary, cosmic-ray flux in particles/cm² sec sr; P_s is the effective cross section for the production of S in meteoritic matter; and N is the number of atoms per gram of meteorite.

The effective cross section, in turn, equals

$$P_s=\int_0^\infty\left[\sum_z \frac{N_z}{N}\sigma_{sz}(E)\right]F(E)dE \Big/ \int_0^\infty F(E)dE, \quad (3)$$

where N_z is the concentration of element Z in the meteorite, in atoms g^{-1}; $\sigma_{sz}(E)$ is the cross section for the production of S from Z at a bombarding energy E; and $F(E)dE$ is the energy spectrum of cosmic radiation *in the meteorite.* As defined, this cross section includes a correction factor for shielding and secondary particles.

In the case of a radioactive nuclide, the amount R produced by a constant flux during time t equals,

$$R=(H_r/\lambda)(1-e^{-\lambda t}), \quad (4)$$

where λ is the decay constant. (The assumption of a constant flux is justified in Sec. 2.) If t is long as compared to the half-life, the exponential approaches zero and R approaches the limiting value H_r/λ. Hence,

$$R=H_r/\lambda \quad (\text{for } t\gg t_{\frac{1}{2}}) \quad (5)$$

and, since for the disintegration rate we have

$$dR/dt\equiv A_r=\lambda R, \quad (5a)$$

---

[1] References refer to the bibliography at the end of the paper.

TABLE I. Ar^{39}, Cl^{36}, and C^{14} in meteorites of known date of fall.

Meteorite	Year of fall	C^{14} (dis/min kg)	Cl^{36} (dis/min kg)	Ar^{39a} (dis/min kg)	$\frac{Ar^{39}}{Cl^{36}}$	Reference
Irons						
Aroos	1959	5.5 ±1.6	18.5±0.5	17.2 ±0.52	0.93	k
				16 ±2		f
		1.85±0.26	20.0±0.4			g
Braunau	1847			19.2 ±0.74		k
Charlotte	1835		25.4±0.9	22.8 ±1.0	0.90	k
N'Goureyma	1900			19.2 ±0.79		k
Pitts	1921			15.9 ±0.5		e
Sikhote-Alin	1947	6.7 ±0.8	9.5±0.4	7.07±0.23	0.74	k, h
				4.42±0.22		e, d
			6.9±0.3	7.0 ±0.3	1.01	i, c
		1.55±0.33	14.4±0.4			g
Treysa	1916		20.2±0.5	17.3 ±0.52	0.86	k, l
				13.3 ±0.6		
Chondrites						
Beardsley[b]	1929	51.2±2.7				j
Bruderheim	1960	55.8±3.0				j
		67 ±6				g
Bruderheim[b]		53.3±5.0				j
Forest City	1890	50 ±5				g
Harleton	1961	37.5±2.4				j
		58 ±6				g
Holbrook	1912	62 ±3				g
Kunashak	1949	71 ±5				g
Modoc	1905	60 ±3				g
New Concord	1860	77 ±4				g
Pantar	1938	48.6±2.8				j
Pultusk	1868	44.2±2.4				j
Richardton	1918	78.0±6.0				g
Walters	1946	53.7±2.5				j

Average Ar^{39}/Cl^{36} in irons: 0.89
Average C^{14} content in chondrites: 48.2 dis/min kg[k] or 65±10[g]

[a] Corrected to time of fall.
[b] Stone phase only.
[c] Davis (1959).
[d] Fireman (1958a).
[e] Fireman and DeFelice (1960a).
[f] Fireman and DeFelice (1960b)
[g] Goel and Kohman (1962).
[h] Kohman and Goel (1961).
[i] Sprenkel (1959).
[j] Suess and Wänke (1961).
[k] Vilcsek and Wänke (1961).
[l] Wänke and Vilcsek (1959).

the steady-state decay rate A_r^0 equals the production rate H_r:

$$A_r^0 = H_r = 4\pi I_0 P_r N. \tag{6}$$

After the fall of the meteorite, it is shielded from cosmic rays by the Earth's atmosphere, and the radioactivity decays with its characteristic half-life. At a time θ after the fall, its disintegration rate A_r^θ equals

$$A_r^\theta = A_r^0 e^{-\lambda\theta}. \tag{7}$$

The term A_r^0 in this equation varies from meteorite to meteorite due to shielding. However, one can calculate it from the observed level of a second radioactivity of longer half-life, e.g., Cl^{36} (308 000 yr) or Al^{26} (740 000 yr). For two radionuclides of similar mass, the cross sections show a similar dependence on particle flux and spectrum. Hence, Eq. (6) shows that the activity ratio at the time of fall is simply equal to the ratio of the two production cross sections: $A_{r1}^0/A_{r2}^0 = P_{r1}/P_{r2}$. This ratio can be determined once and for all in meteorites of known date of fall, and can then be used to calculate A_{r1}^0 from A_{r2}^θ.

Thus far, only Ar^{39} ($t_{\frac{1}{2}} = 325$ yr), C^{14}($t_{\frac{1}{2}} = 5760$ yr), and Cl^{36} have been used to determine terrestrial ages. Table I gives the activity levels of these nuclides in meteorites of known fall date. Evidently, the Ar^{39}/Cl^{36} ratio is reasonably constant with an average value of 0.89. Using this figure, terrestrial ages have been calculated for a number of meteorite "finds" from their Ar^{39} and Cl^{36} contents (Table II). Where the Cl^{36} content was not known, an average value of 15 dis/min kg has been assumed. In the case of C^{14}, A_r^0 was assumed equal to the average C^{14} content of dated falls.

It is interesting that 14 of the 16 iron meteorites dated by this method fall outside its present range. This suggests a fairly slow weathering rate for iron meteorites. In fact, Washington County, for which a fall date of 1400 yr before present has been calculated, is described in the literature (Hey 1953) as a "very fresh disk-shaped mass . . . possibly fell in 1916." For Keen Mountain, which appears to have fallen about 900 yr ago, Henderson and Perry (1958) previously inferred from its fresh appearance that it possibly fell between 1940 and 1950.[2] In fact, the terrestrial-residence time of iron meteorites may approach 10^6 yr in some cases, as indicated by the work of Honda, Shedlovsky, and Arnold (1961). They compared the activity ratios of 0.31-Myr Cl^{36}, 2.7-Myr Be^{10}, 0.74-Myr Al^{26}, 1.27×10^9-yr K^{40}, and $\geqslant$2-Myr Mn^{53} in the Williamstown and Grant iron meteorites. These ratios were close to unity in all cases except Cl^{36}, where a ratio of 0.30 was found. This may well indicate a terrestrial age somewhat greater than one half-life of Cl^{36}, or about 0.4–0.5 Myr. Similarly, Fisher and Schaeffer, (1960) suggest, in discussing the low Cl^{36} content of Casas Grandes, that this meteorite might have been on the earth long enough (i.e., $\sim 10^6$ yr) for the Cl^{36} to have decayed to its present level of 1 dis/min kg.

For stones, one would expect a shorter weathering time. Indeed, only two of the six stone finds in Table II have terrestrial ages significantly greater than zero, but the high ages of these two comes as somewhat of a surprise. Previous estimates of the terrestrial weathering time of chondrites were in the range of 1–100 yr.

Among the problems that can be solved by this method are: (1) correlation of historic reports of meteorite falls with later finds (e.g., the Meissen fall of 1164 or the Grimma fall of 1540–1550 with the Steinbach find of 1751); (2) dating of meteorite craters; (3) dating of prehistoric burials.

2. COSMIC-RAY-EXPOSURE AGES OF METEORITES

It is evident from the structure of meteorites that they did not form as independent entities but were once parts of larger bodies. While residing inside these bodies, the meteorites were effectively shielded from cosmic radiation, since the mean free path for absorption of cosmic-ray primaries in meteoritic matter is only about 150 g/cm². The reduction of the meteorites to meter-

---

[2] Actually, the terrestrial ages of Washington County and Keen Mountain have not been established with certainty, since the Cl^{36} contents were not determined.

sized or smaller fragments, therefore, marked the beginning of intense irradiation by cosmic rays. From a study of stable and radioactive, cosmogenic nuclides in meteorites, it is possible to deduce a "cosmic-ray-exposure age" that is closely related to the breakup date of the meteorite-parent bodies.

The total amount of a cosmogenic nuclide S produced in the meteorite since the time it became a closed system T is found by integrating Eq. (2):

$$S=\int_{-T}^{0} H_s dt. \tag{8}$$

This equation is usually simplified by assuming, as in Sec. 1, that both the cosmic-ray intensity and its spectrum have remained constant in time. This assumption now appears justified, since a number of cosmic-ray-induced radioactivities with half-lives from 16 days to 2.6 Myr were found to occur in meteorites in steady-state proportions (Honda, Shedlovsky, and Arnold 1961; Arnold, Honda, and Lal 1961; Honda and Arnold 1961). We then obtain

$$S=H_s t=4\pi P_s I_0 N t, \tag{9}$$

where t is defined as the *effective* time of irradiation by cosmic rays (the "radiation age" or "exposure age").

One must recognize that the usual interpretation of t as an *age* involves two tacit assumptions. First, it is assumed that the onset of cosmic-ray irradiation was abrupt; i.e., prior to this event the meteorite was buried deep enough in its parent body to be completely shielded from cosmic radiation. This assumption obviously does not hold for material from the uppermost surface layers of the parent body. Second, it is assumed that the meteorite's size and shape did not change during the cosmic-ray-exposure era, so that the amount of shielding at a given point remained constant. Recent work by Vilcsek and Wänke (1961) indicates that this assumption is not justified for at least two iron meteorites (Odessa and Sikhote-Alin) that seem to have undergone multiple breakups.

If P_s is known, Eq. (9) can be solved for t directly. Unfortunately, P_s depends on at least four factors that are generally not well known: (1) the cosmic-ray intensity and spectrum along the meteorite's orbit; (2) shielding correction, which depends on the distance of the sample from the preatmospheric surface of the meteorite; (3) intensity and energy spectrum of secondary particles; and (4) production cross section as a function of energy for all spallation processes leading to a particular cosmogenic nuclide.

Most of these uncertainties cancel if *two* cosmogenic nuclides are measured, one of which is radioactive. Using the subscripts s and r for "stable" and "radioactive," we can write from (4), (5a), and (9)

$$t=\frac{S P_r(1-e^{-\lambda t})}{A_r^0 P_s}, \tag{10}$$

which simplifies to

$$t=\frac{S P_r}{A_r^0 P_s} \tag{10a}$$

if $t_{\frac{1}{2}} \ll t$.

TABLE II. Terrestrial ages of meteorite "finds" calculated from Ar^{39} and C^{14} content.

Meteorite	Cl^{36} (dis/min kg)	Al^{39} or C^{14} (italics) (dis/min kg)	Terr. age (years)[a]	Reference
Irons				
Arispe	6.40 ±0.24	−0.24 ±0.17	≥ 1900	i
Bohumilitz		0.12 ±0.38	≥ 1300	i
Bristol		0.14 ±0.32	≥ 1300	i
Canyon Diablo	8.9 ±0.3	≤ 0.024	≥ 2700	g, b
Carbo	8.1 ±0.4	0.0 ±0.7	≥ 800	i
Carbo		0.02 ±0.09	≥ 1700	d, c
Carbo	3.6 ±0.2	≤ 0.08	≥ 1700	g, b
Casas Grandes	1.0 ±0.1	≤ 0.006	≥ 2300	g, b
Clark Co.		−0.9 ±1.3	≥ 900	i, j
Colomera	18.6 ±0.4	−0.54 ±0.35	≥ 1100	i
Dayton	15.6 ±0.4	−0.10 ±0.30	≥ 1600	i
Grant	12.4 ±1.2	0.02 ±0.09	≥ 1900	d, f
Henbury		−0.26 ±0.31	≥ 1700	i
	8.2 ±0.4	*1.34 ±0.16*	"very young"	e
Keen Mountain		2.15 ±0.33	900	i
Lombard	3.23 ±0.28	−0.02 ±0.29	≥ 760	i
Morradal	24.1 ±0.5	−0.05 ±0.34	≥ 1700	i
Mt. Joy	2.02 ±0.36	0.10 ±0.38	≥ 350	i
Narraburra	6.5 ±0.5	0.02 ±0.43	≥ 900	i
Odessa	6.50 ±0.23	−0.08 ±0.19	≥ 1400	i
	7.5 ±0.2	*0.22 ±0.10*	"fairly young"	e
Odessa	7.8 ±0.5	≤ 0.014	≥ 2900	g, b
San Angelo	6.8 ±0.4	−0.02 ±0.29	≥ 1100	i
São Julião de Moreira	1.84 ±0.35	0.09 ±0.35	≥ 340	i
Sardis	3.5 ±0.2	*0.007 ±0.13*	"old"	e
Smithville	2.84 ±0.38	0.34 ±0.38	≥ 390	i
Tamarugal		0.43 ±0.30	≥ 1200	i
Thunda		0.57 ±0.36	≥ 1100	i
Toluca	10.3 ±0.5	0.20 ±0.21	≥ 1300	i
Washington Co.		0.75 ±0.25	1400	d
Chondrites				
Achilles		*57 ±8*	≤ 5600	e
Coldwater		*34 ±3*	5400 ±1400	e
Dimmit		*37 ±5*	2000 ±2000	h
Hugoton		*31 ±5*	3500 ±2000	h
		42 ±4	3600 ±1500	e
Long Island		*32.5 ±3.5*	3000 ±2000	h
Plainview		*43.2 ±2.9*	≤ 2000	h
		61 ±3	≤ 3800	e
Potter		*3.2 ±2.0*	≥ 20 000	h
		−0.2 ±2.2	≥ 21 000	e
Selma		*29 ±2*	6700 ±1400	e
Woodward Co.		*9.0 ±2.1*	14 000 ±3000	h

[a] For irons of unknown Cl^{36} content, a value of 15 dis/min kg was assumed. For the stones, the average C^{14} contents from Table I were used.
[b] Davis (1959).
[c] Fireman (1958a).
[d] Fireman and DeFelice (1960a)
[e] Goel and Kohman (1962).
[f] Heymann and Schaeffer (1961).
[g] Sprenkel (1959).
[h] Suess and Wänke (1961).
[i] Vilcsek and Wänke (1961).
[j] Wänke and Vilcsek (1959).

The shielding correction cancels in this expression. Moreover, if the cross sections of nuclides R and S show a similar energy dependence, the correction for secondary particles also cancels, leaving just the ratio of the weighted-average cross sections:

$$\sum_z (N_z/N)\sigma_{rz} / \sum_z (N_z/N)\sigma_{s,z}.$$

If both R and S are sufficiently close in mass number so that essentially the same target nuclides contribute to their production, this expression finally simplifies to the ratio of the two laboratory cross sections:

$$t\approx\frac{S\sigma_r}{A_r^0\sigma_s}. \tag{10b}$$

TABLE III. Cosmic-ray-exposure ages of stone meteorites.[a]

Meteorite	Class	Cosmic-ray age (Myr) H^3-He^3	$Ar^{39}-Ar^{38}$	Others	"Gas-retention ages" (Gyr) $K^{40}-Ar^{40}$	$(U,Th)-He^4$
Abee	Cek	13[b]				3.6[b]
Benton	C	20.0[c]			3.8[c]	3.6[c]
Breitscheid	Cg	22.1[d]		30[p,q]	3.3[d]	2.6[d]
Bruderheim	C	24[e]	26[f]	33[r,s]	1.6[e]	1.5[e]
		35[f]		27[r,q]		
Elenovka	Ck	23.6[g]			4.0[g]	4.1[g]
Kandahar	C	22.2[c]			4.3[c]	3.4[c]
Kunashak	Ckab	2.8[g]			0.7[g]	0.4[g]
Mezel	Cia	4.7[c]			3.1[c]	1.3[c]
Monte das Fortes	C	23.6[g]				4.0[g]
Murray	K			15[m]	1.9[t]	
Ramsdorf	Cb	3.4[h]			0.37[h]	0.4[h]
Richardton	Cca		80[m,n]		4.15[o]	3.9[i]
St. Michel	Cw	34[i,j]	110[j,o]		4.0[o]	1.9[i]
Norton Co.	Au	230[l]	500[j]		4.4[o]	≥2.4[i]

[a] Several of the older values were recalculated by Geiss, Oeschger, and Signer (1960) with $\sigma_{\mathrm{H}3}/\sigma_{\mathrm{He}3}/=1$. Where analyses for K, U, and Th were not available, the following average contents were assumed for chondrites: K = 0.85%; U = 11 ppb; Th/U = 3.6 (Edwards and Urey 1955; Hamaguchi, Reed, and Turkevich 1957; Bate, Huizenga, and Potratz 1959). For Ar^{39}/Ar^{38} ages of chondrites, $\sigma_{38}/\sigma_{39}=1.2$ was assumed (Stoenner, Schaeffer and Davis, 1960).

[b] Begemann, Eberhardt, and Hess (1959).

[c] Geiss, Oeschger, and Signer (1960).

[d] Goebel and Schmidlin (1959); König, Wänke, and Mayne (1959); Ebert, Hernegger, König, and Wänke (1959); König and Wänke (1959);

[e] Signer (1961a).

[f] Fireman and DeFelice (1961).

[g] Geiss, Hirt, and Oeschger (1960).

[h] Goebel, Schmidlin, and Zähringer (1959).

[i] Eberhardt and Hess (1960).

[j] Fireman and DeFelice (1960a).

[l] Begemann, Geiss, and Hess (1957).

[m] Stoenner, Schaeffer, and Davis (1960).

[n] Reynolds (1960b).

[o] Geiss and Hess (1958).

[p] Vilcsek and Wänke (1960).

[q] $Na^{22}-Ne^{22}$ (assumed $\sigma_r/\sigma_s=1$).

[r] Honda, Umemoto, and Arnold (1961).

[s] $Cl^{36}-Ar^{36}$ (assumed $\sigma_r/\sigma_s=1.5$).

[t] Stauffer (1961a).

If R and S are an isobaric pair (e.g., H^3-He^3, Cl^{36}-Ar^{36}), the nuclide S is produced by decay of R as well as directly, and we, therefore, write

$$t\approx\frac{S\sigma_r}{A_r^0(\sigma_r+\sigma_s)}. \qquad (10c)$$

To date, two isobaric pairs (H^3-He^3 and Cl^{36}-Ar^{36}) and two isotopic pairs (Ar^{39}-Ar^{38} and K^{40}-K^{41}) have been used for age determinations. In addition, several less-direct methods have been employed. In some cases, stable nuclides were used that were rather far in mass number from the radioactive nuclide, e.g., Ar^{39}-He^3 (Fireman and DeFelice 1960a) or Ar^{39}-Sc^{45} (Wänke and Vilcsek 1959). The relative production cross sections were taken from laboratory determinations where available or from spallation theory (Miller and Hudis 1959; Rudstam 1955).

Goel (1960) has attempted to calculate production rates in iron meteorites for all nuclides with $17\leq Z\leq 25$ and $34\leq A\leq 54$, as well as H^3, He^3, and He^4. His calculations are based on cosmic-ray star-production rates and on the semi-empirical, spallation-yield curves of Rudstam (1955). He suggested that the relative production rates of any two nuclides should remain approximately constant for about three interaction lengths (~70 cm in iron and ~120 cm in stone). Hence, his calculated values should be applicable to meteorites of this radius or smaller and to the surface regions of larger meteorites. This includes nearly all cases of practical importance, since radioactivity levels in the interiors of large meteorites are likely to be too low to be conveniently measurable.

Still another approach has been used by Fisher and Schaeffer (1960). They pointed out that each of the three ratios Ne^{21}/Ar^{38}, He^3/Ar^{38}, and He^3/He^4 was depth dependent, thus providing a basis for estimating the degree of shielding. While any one of these ratios might be anomalous due to some compositional peculiarity (e.g., trapped primordial He^4; production of Ne^{21} from Mg, etc.), comparison of all three ratios would permit identification and rejection of the anomalous one.

In this manner, Fisher and Schaeffer ranked 15 meteorites on a relative scale in the order of increasing depth and decreasing production rate of cosmogenic nuclides. This relative scale was then converted to an absolute one by relating it to the Cl^{36} measurements of Sprenkel (1959) for six of the above meteorites. The meteorites were then divided into three groups of approximately equal degree of shielding. For each group, the production rates for He^3, Ne^{21}, and Ar^{38} were estimated from the average Cl^{36} content and the estimated ratio of the production cross sections. From these production rates, three ages were calculated for each meteorite. The average of these ages ("modified Cl^{36} age") was expected to be more reliable than the individual ages. Unfortunately, the validity of this method has become open to doubt. Wänke (1960a) pointed out that, in view of the similarity in energy dependence of the cross sections of He^3 and Ar^{38}, the ratio of these nuclides should show little if any depth variation. Although Schaeffer (1960) maintains that, even in the absence of a theoretical justification, it is still possible to use the empirically observed, strong variation of the He^3/Ar^{38} ratio for dating purposes, Signer and Nier (1961) found only a slight variation in

He^3/Ar^{38} ratio among 19 iron meteorites. A further source of uncertainty comes from the Cl^{36} measurements used by Fisher and Schaeffer. As pointed out in Sec. 1, the low Cl^{36} content of Williamstown is not so much due to shielding as to decay since fall. The same appears to be true of Carbo.

Another approach to the problem of estimating production rates has been developed by Singer (1952), by Martin (1953), by Ebert and Wänke (1957), and by Hoffman and Nier (1958). The production rate of a given nuclide at a given depth below the surface can be written as the sum of two separate functions for primary and secondary particles. If two nuclides are chosen whose production cross sections differ in energy dependence, e.g., He^3 and He^4, and their concentration is measured at a number of points in the meteorite, then all parameters in the equations can be obtained by curve fitting. Thus, it is possible to obtain not only the production rates of He^3 and He^4 at any point in the meteorite, but also its preatmospheric radius. Although the ages thus calculated ("He^3-He^4 ages") are obtained by a less direct route than those involving the measurement of a radionuclide, this method is capable of giving results of rather high accuracy.

A further refinement of this method was developed by Signer and Nier (1960, 1961). The concentration of a given cosmogenic nuclide is a function of three unknowns: size of the meteorite, shielding depth, and exposure age. If four cosmogenic nuclides are measured (He^3, He^4, Ne^{21}, and Ar^{38}), the problem becomes mathematically overdetermined. It is thus possible to calculate three exposure ages for each meteorite. The degree of concordance of these values serves as a check on the validity of the model and the calculation.

Another method of great potential value, based on cosmogenic $K^{40}(t_{\frac{1}{2}}=1.27\times10^9$ yr), was proposed by Voshage and Hintenberger (1959, 1961). They developed a technique for isolating virtually pure cosmogenic potassium from iron meteorites, and noted that its $K^{39}/K^{40}/K^{41}$ ratio (42.0/18.9/39.1 in a typical case) differed radically from terrestrial potassium (93.08/0.0112/6.91). Since potassium has *two* stable isotopes (K^{39} and K^{41}), the measured mass spectrum could be corrected for terrestrial (or preterrestrial) contamination with normal potassium. The exposure age t of the meteorite is then related to the observed isotopic ratios by the equation

$$\frac{\lambda t}{1-e^{-\lambda t}}=\frac{(K^{41}/K^{40})-a(K^{39}/K^{40})}{(P_{41}/P_{40})-a(P_{39}/P_{40})}=\frac{M}{N},$$

where λ is the decay constant of K^{40}, P is the production rate, and a is the K^{41}/K^{39} ratio in normal potassium. The numerator M, containing the observed isotopic ratios, depends on the exposure age, the degree of shielding, and the chemical composition. To cancel the effect of the last two variables, the production rates in the denominator N must be known. Voshage and Hintenberger (1961) obtained an approximate value of N for each meteorite by comparing the observed He/Ne ratio with the theoretical production rates for He, Ne, and the three potassium isotopes calculated according to Arnold, Honda, and Lal 1961. The agreement between this method and others is still somewhat unsatisfactory, but is likely to improve once the dependence of N on shielding depth has been experimentally determined.

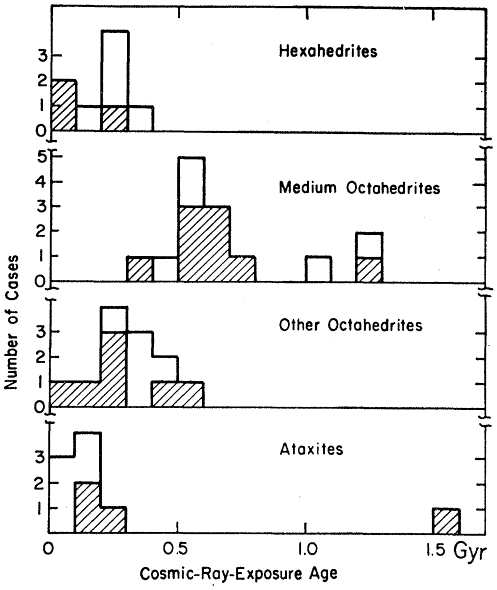

FIG. 1. Cosmic-ray-exposure ages of iron meteorites. (Data from Table IV). Shaded area = selected values of somewhat greater reliability ("reviewer's guess" in Table IV). Note contrast between hexahedrites and medium octahedrites.

2.1 Experimental Data

Tables III and IV contain a summary of cosmic-ray-exposure ages of stone and iron meteorites. Only those published ages are listed that include a shielding correction. Data for iron meteorites are also plotted in Fig. 1. In the case of stone meteorites, the "gas-retention" ages based on radiogenic Ar^{40} and He^4 (Sec. 5) are also tabulated for comparison. Before going into a detailed interpretation of the data, it is necessary to consider their reliability.

The relative values of the H^3-He^3 ages of the stones are generally quite well determined (about $\pm10\%$), though the absolute values are rather less well known. In those few cases where intercomparison between the H^3-He^3 and Ar^{39}-Ar^{38} methods is possible, the agreement is far from satisfactory. This discrepancy may in large part be due to our ignorance of the true production cross sections. For example, the relative cross sections for the production of H^3 and He^3 are determined from iron targets exposed to monoenergetic proton beams. The cosmic radiation, on the other hand, consists of particles varying enormously in energy; in addition,

TABLE IV. Cosmic-ray-exposure ages of iron and stony-iron meteorites (Gyr).[a]

Name	Class	$Ar^{39}-X$	$Cl^{36}-Ne^{21}$	$Cl^{36}-Ar^{36}$	Indirect Cl^{36}	$He^{3}-X$	$K^{40}-X$	Reviewer's guess
Arispe	Ogg		*0.44*^{d}		0.35^{m}			
Aroos	Og	$0.52^{b,c}$	0.54^{d}	0.53^{b}		$0.8^{t,u}$	$0.94^{z,aa}$	0.53
Braunau	H	$0.008^{d,e}$						0.008
Bristol	Of						$0.48^{z,aa}$	
Canyon Diablo	Og			0.16^{m} 0.18^{h}				0.17
Carbo	Om		0.72^{d}	*1.2*^{m} *1.2*^{n} *1.42*^{h}	*1.1*^{m}	$0.9^{v,w}$ $0.6^{t,u}$	*1.2*^{e,aa} $0.895^{z,aa}$	0.7
Casas Grandes	Om			1.0^{m}	0.109^{m}	$0.7^{v,w}$ $0.25^{t,u}$		
Charcas	Om					$0.6^{t,u}$		0.6
Charlotte	Of	$0.25^{d,e}$					$0.315^{z,aa}$	0.25
Clark County	Om					*4.8*^{x,w} $1.0^{t,u}$	$1.40^{z,aa}$	1.2
Coya Norte	H					$0.25^{t,u}$		
Dayton	D_1		0.14^{d}					0.14
Deep Springs	D_1					*2.9*^{x,w} $1.5^{t,u}$		1.5
Forsyth County	D_2				0.075^{m}			
Grant	Of			0.59^{b}	*2.00*^{r}	$0.6^{v,w}$	*1.98*^{h,cc}	0.59
Henbury	Om				1.2^{s}			
Keen Mountain	H	$0.16^{d,e}$				$0.2^{v,w}$ $0.2^{t,u}$		0.2
Lombard	H		*0.26*^{d}				$0.205^{z,aa}$	
Merceditas	Om					$0.6^{t,u}$		0.6
Morradal	D_1		0.12^{d}					0.12
Mt. Joy	H		*0.35*^{d}					
Narraburra	Om		*0.47*^{d}					
Negrillos	H					$0.03^{t,u}$		0.03
N'Goureyma	Obzg	$0.235^{d,e}$						0.24
Norfork	Om			0.56^{b}		$0.56^{h,y}$		0.56
Odessa	Og		*0.024*^{d,q} *0.085*^{d,q}	0.37^{m} 0.42^{h}	0.325^{m}	$0.45^{t,u}$	$0.42^{h,dd}$	0.4
Pará de Minas	Om					$1.05^{h,y}$		
Pitts	Obc	$0.03^{f,g}$ *0.082*^{h,g}						0.03
Ria Loa	H					$0.15^{t,u}$		
San Angelo	Om	$0.48^{d,e}$						
Santa Catharina	D_1				0.166^{m}			
Santa Rosa	D_2-Ob				0.140^{m}			
São Julião de Moreira	Ogg		*0.27*^{d}					
Sikhote-Alin	Ogg	$0.22^{f,i,g}$ *1.04*^{h,g} $0.25^{b,c}$	0.06^{d}	0.24^{b} 0.17^{m} 0.19^{h}	0.165^{m}	$0.3^{t,u}$		0.22
Smithland	D_1				0.090^{m}			
Smithville	Og		*0.32*^{d}			$0.515^{z,aa}$		
Tamarugal	Om							
Tocopilla	H				0.072^{s}	$0.25^{t,u}$		
Toluca	Om		0.60^{d}	0.46^{h}	*0.081*^{m} *0.11*^{s}	$0.25^{t,u}$		0.5
Treysa	Om	$0.35^{f,i,g}$ $1.07^{h,g}$ $0.30^{j,k,c}$ $0.29^{b,c}$ $0.6^{j,l}$	0.45^{d}	$0.30^{j,k}$	*1.50*^{s} *1.3*^{i}	*1.14*^{h,y}	*0.59*^{e,aa} $0.61^{z,aa}$	0.31
Tucson	Dm				0.020^{m}			
Washington County	D_1				0.285^{m}	$0.2^{t,u}$		0.2
Williamstown	Om			*2.2*^{m} *2.45*^{h}	*1.72*^{m}	$0.65^{e,u}$	*2.10*^{h,cc} $0.725^{z,aa}$	0.65
Admire	P			$0.14^{ce,p}$		$0.15^{t,u}$		0.14
Colomera	P		0.075^{d}					0.075
Imilac	P				0.74^{s}			

[a] The ages that are italicized are believed to be in error. This selection was based on the following criteria;

1. Goel's $Ar^{39}-He^{3}$ ages for Sikhote-Alin and Treysa recalculated from the data of Fireman and De Felice are much higher than their original values which, in turn, are somewhat higher than the well-determined $Cl^{36}-Ar^{36}$ age for Sikhote-Alin (Heymann and Schaeffer 1961) and the relatively reliable $Ar^{39}-Ar^{38}$ age for Treysa. Goel's $Ar^{39}-He^{3}$ age for Pitts is, therefore, also likely to be in error.

2. The $Cl^{36}-Ne^{21}$ ages of Vilcsek and Wänke (1961) are based on the tacit assumption that the two cross sections have identical energy dependence, in spite of the large difference in mass number. This is in disagreement with theory and with the experimental data of Signer and Nier (1960) who found a strong depth variation of the Ne^{21}/Ar^{38} ratio in Grant. For this reason, all ages are considered suspect that are based on Cl^{36} contents less than 10 dis/min kg (compare with the maximum observed value of 25.4 dis/min kg for Charlotte). This criterion also serves to eliminate

it contains some 10 to 20% helium-4 nuclei which may become converted to He^3 or H^3 by stripping reactions. These factors could change appreciably the relative production rates of H^3 and He^3 in the meteorite (Singer 1958, 1960). Similarly, the ratio of the laboratory cross sections for production of Ar^{38} and Ar^{39} in iron is 2 (Schaeffer and Zähringer 1959). In a stone meteorite containing substantial amounts of calcium and potassium, a large part of the argon may be made from these elements rather than from iron. In that case, the proximity in mass number of target and residual nucleus becomes of importance, and reactions such as $Ca^{40}(p,3p)Ar^{38}$ may shift the ratio heavily in favor of Ar^{38}. The situation is further complicated by meson and α-particle reactions. However, all these difficulties affect primarily the determination of *absolute* ages. It should still be possible to obtain an accurate *relative* age scale by measuring meteorites of the same composition by the same method.

In some iron meteorites (e.g., Sikhote-Alin, Aroos), tritium is lost *after* the fall in some mysterious way, presumably by diffusion (Fireman and DeFelice 1960; Geiss, Hirt, and Oeschger 1960; Bainbridge, Suess, and Wänke 1961). A partial loss may have occurred in other irons, and even in the metal phase of chondrites. Geiss, Hirt, and Oeschger (1960) found the specific activity of H^3 in the metal phase of Elenovka to be about 70% that in the whole meteorite and attributed this difference mainly to the lower H^3-production cross section in metal. On the other hand, Fireman and DeFelice (1961) found this value to be only 35% in Bruderheim, and considered this an indication of diffusion loss. Until this matter is clarified, it would be well to treat with reserve all H^3 ages of iron meteorites.

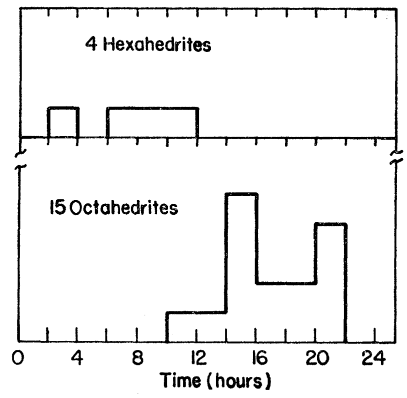

FIG. 2. Time of fall of iron meteorites. (Wänke 1960a).

2.2 Interpretation of the Data

The picture offered by these ages is still somewhat confusing, though certain regularities are beginning to appear. The following trends can be inferred from Tables III and IV:

(1) At least 6 of the 13 chondrites in Table III have virtually the same H^3-He^3 age.

(2) For three other chondrites, there appears to be a correlation between short H^3-He^3 age and short K^{40}-Ar^{40} and (U,Th)-He^4 ages.

(3) The ages of the irons, as a group, tend to run considerably higher than those of the stones.

(4) The ages of the irons show a tendency to cluster. In a set of 5 meteorites that were dated with particularly high accuracy by the Cl^{36}-Ar^{36} and Ar^{39}-Ar^{38} methods, three (Aroos, Grant, Norfork) have ages of 0.50–0.59×10^9 yr, while two others (Sikhote-Alin and Treysa) have ages of 0.24–0.29×10^9 yr (Heymann and Schaeffer 1961). In the three cases where the same meteorite was dated by both methods, the agreement was excellent. Of 19 iron meteorites dated by Signer and Nier (1961), 16 lie near one or the other of these two values.

(5) Some correlations between age and structural class may be present. The medium octahedrites appear to be uniformly older than 0.3×10^9 yr, whereas most hexahedrites and nickel-rich ataxites have ages of less than 0.3×10^9 yr (Vilcsek and Wänke 1961). Moreover, Wänke (1960b) has drawn attention to the fact that all known hexahedrite falls occurred between midnight and noon, whereas the octahedrites, with one exception, fell between noon and midnight (Fig. 2).

It is instructive to review the principal hypotheses on the subject and to examine their consistency with the above data.

---

meteorites with long terrestrial ages, in which the Cl^{36} level has been reduced by decay.

3. The $Cl^{36}-Ar^{36}$ and modified Cl^{36} ages of Carbo and Williamstown are apparently high due to decay of Cl^{36} since their fall.

4. The indirect Cl^{36} ages of Toluca and Treysa disagree with two Cl^{36} ages and three Ar^{39} ages, respectively.

5. The H^3-He^3 age of Treysa is in doubt because of possible tritium losses (Fireman and DeFelice 1960).

6. Bauer's ages for Clark County and Deep Springs are based on the He^3/He^4 ratio in a single sample. The absolute ages of these meteorites were calculated relative to Williamstown = 2.2 Gyr, a value now known to be high.

7. The $K^{40}-K^{41}$ age of Treysa disagrees with three Ar^{39} ages. The $K^{40}-K^{41}$ age of Carbo, based on the same assumed cross sections, is also likely to be high.

8. The $K^{40}-Ar^{36}$ age of Grant disagrees with the well-determined $Cl^{36}-Ar^{36}$ age and the virtually identical He^3-He^4 as given by Hoffman and Nier (1960). The $K^{40}-Ar^{36}$ age of Williamstown, based on the same assumed cross sections, is, therefore, considered suspect.

[b] Heymann and Schaeffer (1961).
[c] $X = Ar^{38}$.
[d] Vilcsek and Wänke (1961).
[e] $X = Ne^{21}$.
[f] Fireman and DeFelice (1960).
[g] $X = He^3$.
[h] Goel (1960).
[i] Fisher (1961b).
[j] Wänke and Vilcsek (1959).
[k] Fechtig, Gentner, and Kistner (1960).
[l] $X = Sc^{45}$.
[m] Fisher and Schaeffer (1960).
[n] Sprenkel (1959).
[p] The $Al^{26}-Ne^{22}$ age for the stone phase is 0.12 ± 0.02.
[q] These values refer to two different specimens.
[r] Fisher (1961c).
[s] Schaeffer and Fisher (1960).
[t] Signer and Nier (1961).
[u] $X = He^4, Ne^{21}, Ar^{38}$.
[v] Hoffman and Nier (1960).
[w] $X = He^4$.
[x] Bauer (1960, 1961).
[y] $X = H^3$.
[z] Voshage and Hintenberger (1961).
[aa] $X = K^{41}$.
[bb] Marshall, (1959, 1960), Voshage and Hintenberger (1960).
[cc] $X = Ar^{36}$.
[dd] $X = Cl^{36}$.
[ee] Honda, Umemoto, and Arnold (1961).

Paneth and his school, working under the mistaken impression that much of the helium in meteorites was of radiogenic rather than cosmogenic origin, calculated (U,Th)-He^4 ages for a number of meteorites and concluded that the parent body of these meteorites must have disrupted $\sim 2\times 10^8$ years "ago, shortly after the solidification of the metal phase" (Dalton, Paneth, Reasbeck, Thomson, and Mayne 1953; Reasbeck and Mayne 1955). This concept of a single parent body and a single breakup became widely accepted, and many workers have made the additional assumption that all meteorites should, therefore, have the same cosmic-ray-exposure age. However, any meteorites produced by secondary collisions of larger fragments would, of course, have shorter ages. The age spectrum corresponding to a single breakup should, therefore, have a peak at some maximum value and a continuum extending toward lower values. The observed distribution is not in good accord with this prediction and, while this in itself may not be a strong argument against the hypothesis of a single breakup, other more compelling arguments can be raised against it (Anders and Goles 1961).

A second view is that the meteorites are produced more or less continuously by collisions in the asteroidal belt. Kuiper (1950) has estimated a collision probability of 0.1 in 3×10^9 yr for any two of the largest 10 asteroids alone, and Piotrowski (1953) calculated collision lifetimes of 10^8–10^9 years for typical asteroids. With some 30 000 observable asteroids, one would expect catastrophic collisions at intervals of 10^4–10^5 yr, i.e., virtually continuously. Since the collision cross section per unit mass increases with decreasing radius, one would expect secondary collisions among fragments to damp out any fluctuations in the meteorite-production rate due to primary collisions among asteroids (Kuiper 1950).

That most asteroids have indeed been affected by collisions is attested to by the variability in their light curves (Groeneveld and Kuiper 1954). The hypothesis of continuous meteorite production by asteroidal collisions would, therefore, be perfectly acceptable, were it not for the curious, systematic difference between the exposure ages of stones and irons.

On the basis of the data available in 1959 (most of which were, however, calculated without allowance for shielding), it appeared that the stones had ages typically the order of 20–30 Myr, whereas the irons had ages of 10^8–10^9 years. Urey (1959) pointed out that these figures happened to be of the same order as the collision lifetimes of objects coming from the Moon and the asteroidal belt (Öpik 1951). He, therefore, proposed that the iron meteorites (and a few stones with high ages, such as Norton County) came from the asteroidal belt, whereas the stones (and a few irons, such as Horse Creek) were spalled off the Moon by impact of meteorites from the asteroidal belt.

Urey's idea has been criticized by Goles, Fish, and Anders (1960) as being incompatible with the high K^{40}-Ar^{40} ages of the chondrites, though no better explanation of the differences in exposure ages was offered. On the other hand, Fireman and DeFelice (1960a) and Whipple and Fireman (1959) have suggested "space erosion" by interplanetary dust as the cause of these differences.

The apparent age of T_E of material in the center of a spherical meteorite of true age T equals (Whipple and Fireman 1959; Eberhardt and Geiss 1960a):

$$T_E = (L/E)(1 - e^{-(E/L)T}), \tag{11}$$

where L is the absorption length of cosmic radiation in the meteorite and E is the erosion rate. The limiting cases of this equation are (Eberhardt and Geiss 1960a)

$$T_E = T \quad \text{for} \quad T \ll L/E \text{ (no space erosion)}, \tag{11a}$$

$$T_E = L/E \quad \text{for} \quad T \gg L/E \text{ (space erosion dominant)}. \tag{11b}$$

The erosion rate calculated from Öpik's theory of meteor impact (Öpik 1958) is

$$E = \frac{1.12 P w_0^2}{(s/\rho)^{\frac{1}{2}}}, \tag{12}$$

where P is the density of interplanetary dust, w_0 is the impact velocity, s is the crushing strength, and ρ is the density of the meteoritic body.

Fireman and DeFelice argue that stones of low crushing strength (e.g., the spherical chondrites, $s \approx 3\times 10^6$ dynes/cm^2) should erode much faster than harder stones ($s \geq 3\times 10^9$ dynes/cm^2) and presumably irons ($s \sim 3\times 10^9$ dynes/cm^2, Buddhue 1941). The erosion rates calculated for $w_0 = 10$ km/sec and $P = 5\times 10^{-21}$ g/cm^3 (van deHulst 1947) are 200 g cm^{-2} (Myr)$^{-1}$ for spherical chondrites and 5 g cm^{-2} (Myr)$^{-1}$ for the hardest stones and irons. At such high erosion rates, apparent ages of 10^6 yr and 5×10^7 yr would be expected for stones and irons.

The disagreement in the absolute values is not serious and could be eliminated by a different choice of parameters. However, as Eberhardt and Hess (1960), Eberhardt and Geiss (1960a), and others have pointed out, there are serious qualitative inconsistencies with the data. Abee, a very hard stone, has an age of 13 Myr, whereas the friable Norton County has an age of 230 Myr. Moreover, it would seem that the parameter of importance is not the *macroscopic* crushing strength but the strength of the meteoritic minerals, since a micron-sized dust particle impacting with supersonic velocities interacts only with a small fraction of a crystal grain and not with the entire meteorite. The mechanical strength of silicates does not differ much from that of iron and, even allowing for some second-order effects (such as the greater ductility of the metal), the erosion rates should not be greatly different. This applies also to two other erosion mechanisms discussed by Fireman and DeFelice: particle radiation from the sun and expanding, coronal gases.

When the impacting particles approach or exceed the size of the individual crystal grains in the meteorite, the erosion rate will indeed become dependent on the crushing strength, or brittleness, of the meteorite (Eberhardt and Hess 1960; Fisher 1961a). Low-velocity collisions among the debris in the asteroidal belt must be rather frequent, and some degree of erosion undoubtedly takes place by this mechanism. But again, there seems to be little correlation between brittleness and the observed ages.

The preceding discussion has ignored two important trends of the data in Tables III and IV: the clustering of ages for the chondrites and the apparent correlation between age and structural class for the irons. These trends change the picture rather drastically, but since they are still somewhat tentative and, moreover, were not known when the principal interpretations of cosmic-ray-exposure ages were formulated, they are discussed separately in the following.

It is certainly striking that at least 6 of the 13 chondrites in Table III have virtually the same H^3-He^3 age, 22±2 Myr. Geiss and Oeschger (1960), therefore, suggested that these chondrites were produced in a single breakup process about 22 Myr ago. Of the remaining seven chondrites, three have very short H^3-He^3 ages, possibly corresponding to another breakup. However, since their K^{40}-Ar^{40} and (U,Th)-He^4 ages are also short as compared to those of the other chondrites, it is not unlikely that they suffered diffusion losses of noble gases due to close approaches to the sun (see Sec. 5.3). Their true cosmic-ray-exposure ages may, therefore, be somewhat greater than the 3–5 Myr indicated by the data.

Two other meteorites, Abee and Murray, belong to two mineralogically and chemically distinct subclasses that may well have originated in separate bodies, and the last two, Richardton and St. Michel, have ages that are not inconsistent with a true value of 22 Myr. However, there undoubtedly are stone meteorites that have decidedly longer or shorter ages, judging from their He^3 content, e.g., Beardsley (9 Myr, Eberhardt and Hess 1960).

For the irons, only a few precision ages are available, and the systematic errors of most are still too large to permit an intercomparison of data obtained by different methods or different investigators. However, as pointed out above, there appears to be a tendency for the ages of many irons to cluster in the regions 0.25–0.3 and 0.5–0.6×10^9 yr. Also, some correlation seems to exist between age (and fall time) and structural class, at least for the hexahedrites and medium octahedrites.

It is thus necessary to account for at least the chondrite cluster at 22 Myr, and possibly also for the iron clusters at 0.25–0.3 and 0.5–0.6×10^9 Myr. These facts are not easily explained by any of the existing theories.

Breakup of a single planet would, as already pointed out, give a single, large cluster and a continuum of shorter ages. This is not in accord with the data.

Collisions of asteroids on a time scale of 10^4 to 10^5 yrs would give a continuum of ages with some fluctuations due to differences in the size and relative velocity of the colliding bodies. Clustering of chondrite ages at 22 Myr could be attributed simply to a collision of larger-than-average bodies. However, this explanation is inadequate if the clustering of the irons is taken into account. It is certainly puzzling that the first cluster is limited to chondrites and the second to irons. One can, of course, assume that the parent bodies of these meteorities differed in composition, being of stone in one case and of iron in the other. For bodies derived from a dust cloud with comparable abundances of Fe and Si, such extreme variations in composition are very unlikely. One should, therefore, consider the alternative possibility that the collision itself produces some compositional sorting of the material. To become a meteorite datable by Earth men, a fragment must be thrown into an orbit with a perihelion of ≤ 1 a.u. This requires an impulse of the right magnitude and direction. Our understanding of the dynamics of asteroidal collisions is very limited, but it is highly probable by analogy with meteorite impact that the velocity spectrum of the fragments will vary with distance from the impact focus (Öpik 1958, Shoemaker 1960, and Bjork 1961). Just which fragments from which regions of the colliding bodies will actually be thrown into the required, highly eccentric orbits will depend on the impact parameters. These will vary from case to case, and if we assume that the various classes of meteorites come from successive layers of differentiated asteroids, it is not difficult to see, by hindsight, how only a small fraction of the material, representing a narrow range in composition, may be placed in meteoritic orbits.

The clustering of ages does not contradict Urey's (1959) theory, though it weakens some of the premises on which it was based. If the meteorites were produced in a very small number of collisions, then any age differences between irons and stones must be regarded as accidental rather than systematic. At some other era in the Earth's history, exactly the opposite situation might prevail. The problem still remains as to why some collisions seem to produce only stones and others only irons. The easiest way to account for this puzzle would be to postulate bodies of different chemical composition. Indeed, Urey had argued earlier that the achondrites and irons came from "primary" objects of lunar size and the chondrites from "secondary" objects of asteroidal size (Urey 1957a, 1958). This is certainly consistent with the chondrite cluster at 22 Myr, but would lead one to expect two clusters of achondrites at 0.25–0.3 and 0.5–0.6×10^9 yr, complementary to the iron clusters. Correlations of this type should be looked for in future work.

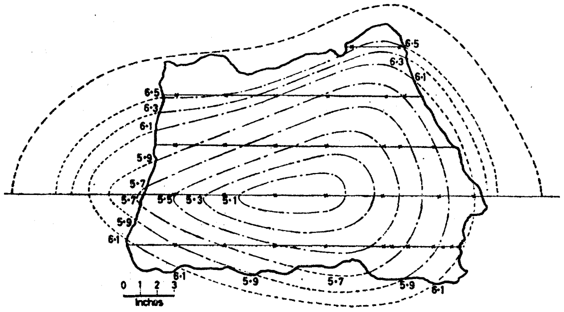

FIG. 3. Contours of constant He^3 content in Grant meteorite (Fireman 1958).

The space-erosion theory is perhaps least able to account for the clustering of ages. For 6 of 13 chondrites to have an age of 22±2 Myr requires that they were exposed to precisely the same degree of erosion, regardless of when they originated. The density of interplanetary dust varies with distance from the sun and from the ecliptic plane, so that the degree of erosion varies with the orbital elements, particularly the inclination. The 6 meteorites in question fell on different days of the year, and were, therefore, in different orbits. Hence, it is difficult to see why they should have been eroding at exactly the same rate. It thus seems that space erosion must have been a minor, rather than a major, factor in determining the cosmic-ray-exposure ages of meteorites.

3. PREATMOSPHERIC SIZE OF METEORITES

Still another problem that has been elucidated by the study of cosmogenic nuclides is the preatmospheric size and shape of meteorites (Singer 1952, 1954). Owing to the absorption of cosmic rays, the concentration of a given cosmogenic nuclide will decrease with distance from the preatmospheric surface. By measuring the concentration of such a nuclide, e.g., helium, at a number of points in a meteorite slice, it is possible to construct contours of equal helium content. These should run parallel to the original preatmospheric surface.

Fireman (1959) has attempted to reconstruct the original shape of Grant from measured contours of He^3 (Fig. 3). He was able to show that ablation was not uniform and calculated a preatmospheric mass of 880 kg. The postatmospheric mass is 480 kg, so that a mass loss of about 45% seems to have taken place.

The same meteorite was also studied by Hoffman and Nier who measured both He^3 and He^4. Their results are in tolerable agreement with Fireman's, though they differ in details (Fig. 4). Treating the meteorite as spherical, they computed the ablation loss from the theoretical depth dependence of the He^3/He^4 ratio, and found a ratio of postatmospheric to preatmospheric radius of 0.65, corresponding to a 73% mass loss.

This is rather higher than the values derived from a metallographic determination of the ablation rate for Grant (0.1–0.2 cm/sec; Maringer and Manning 1960).

TABLE V. Ablation losses of iron meteorites.

Meteorite	Postatmospheric mass (kg)	Ablation loss %	Method	Reference
Carbo	~450	92	He^3/He^4	Hoffman and Nier (1959)
		78	He/Ne	Ebert and Wänke (1957)
Casas Grandes	1550	86	He^3/He^4	Hoffman and Nier (1960)
Grant	480	73	He^3/He^4	Hoffman and Nier (1958)
		45	He^3	Fireman (1959)
		20–48	Metallogr.	Maringer and Manning (1960)
Keen Mountain	6.75	99	He^3/He^4	Hoffman and Nier (1960)
Mt. Ayliff	~14	78	He/Ne	Ebert and Wänke (1957)
Tawallah Valley	75.75	27[a]	Metallogr.	Lovering, Parry, and Jaeger (1960)
Wedderburn	0.2	60[a]	Metallogr.	Lovering, Parry, and Jaeger (1960)

[a] Although Lovering *et al.* and Maringer and Manning find virtually identical ablation *rates* (0.18 and 0.1–0.2 cm/sec), their results are not directly comparable, since their estimates for the ablation *time* differ by nearly a factor of 10 (3.3 sec vs 20–30 sec).

At ablation times of 20–30 sec, the total thickness ablated is 2–6 cm, corresponding to mass losses of 20–48%. But it must be noted that the metallographic method is likely to underestimate ablation losses, since it only measures the *final* ablation rate. The record of earlier, higher ablation rates is, of course, destroyed by subsequent ablation.

Three other meteorites have been studied by this technique thus far: Carbo (Fireman 1958b, Hoffman and Nier 1959), Keen Mountain, and Casas Grandes (Hoffman and Nier 1960). Carbo and Casas Grandes, in particular, seem to have broken up after their entry into the atmosphere (Fig. 5). Estimates of the mass loss for other meteorites have been made on the basis of metallographically determined ablation rates, or the He/Ne ratio. The results are given in Table V.

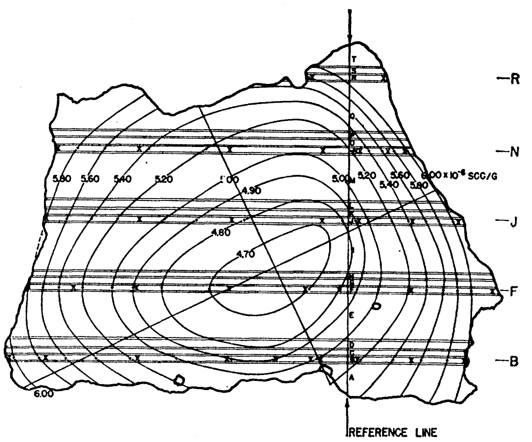

FIG. 4. Contours of constant He^3 content in Grant meteorite (Hoffman and Nier 1958). Compare with Fig. 3.

The ablation loss of a meteorite depends strongly on its geocentric velocity (Thomas and Whipple 1951, Martin 1953) and one can, therefore, expect large variations in the mass losses. For this reason alone, none of the data in Table V can be rejected outright, though several of the values are somewhat doubtful. For example, the high mass loss of the Keen Mountain hexahedrite was inferred from its low $\mathrm{He}^3/\mathrm{He}^4$ ratio, which may, however, be due to tritium diffusion loss.

4. SOLIDIFICATION AGES

Some time after the formation of the meteorite parent bodies, they underwent extensive melting, leading to a partial separation of metal and silicate phases and other chemical fractionations. The formation of the parent bodies cannot be dated by radioactivity, but the melting and fractionation processes can.

For these dating methods, the following long-lived radioactivities are available:

$$\mathrm{Rb}^{87}\xrightarrow[47\text{ Gyr}]{\beta^-}\mathrm{Sr}^{87},$$

$$\mathrm{Re}^{187}\xrightarrow[43\text{ Gyr}]{\beta^-}\mathrm{Os}^{187},$$

$$\mathrm{U}^{235}\xrightarrow[0.71\text{ Gyr}]{7\alpha}\mathrm{Pb}^{207},$$

$$\mathrm{U}^{238}\xrightarrow[4.51\text{ Gyr}]{8\alpha}\mathrm{Pb}^{206},$$

$$\mathrm{Th}^{232}\xrightarrow[13.9\text{ Gyr}]{6\alpha}\mathrm{Pb}^{208}.$$

For practical reasons, no use has been made of the decay of Th^{232} to Pb^{208}. Also, the data for the two uranium decay chains are generally combined in the $\mathrm{Pb}^{207}/\mathrm{Pb}^{206}$ method.

All these methods date a chemical fractionation between parent and daughter element. The ideal case is represented by a mineral that initially contained the parent element only, so that all of the daughter element present can be attributed to radioactive decay. Unfortunately, the chemical fractionations in meteorites are not only less diverse and less complete than in the earth, but the fractionation, if any, often tends to go in the wrong direction, leading to an enrichment of the daughter rather than the parent element.

The problems thus posed are rarely encountered in terrestrial geologic dating, and must, therefore, be discussed in some detail. The discussion is given specifically for the Rb^{87}-Sr^{87} chain, although the conclusions apply to the remaining methods as well.

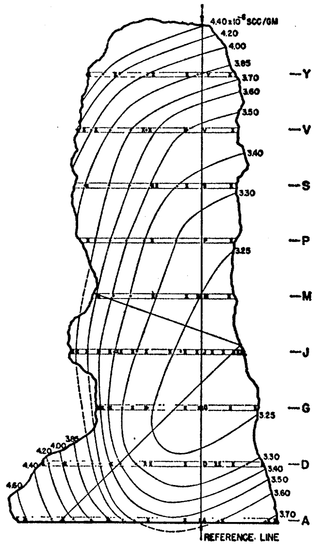

FIG. 5. Contours of constant He^3 content in Carbo meteorite (Hoffman and Nier 1959). Open contours on right-hand side indicate fragmentation late in meteorite's history. Cavity at lower left seems to have formed *after* cosmic-ray irradiation, presumably during entry into Earth's atmosphere.

4.1 Rubidium-Strontium Ages[3]

Consider a meteorite that contains a certain Rb-Sr ratio and became a closed system at time t_0. At some later time t, its content of radiogenic Sr^{87} equals

$$(\mathrm{Sr}^{87})_{r,t}=(\mathrm{Rb}^{87})_t(e^{\lambda\Delta t}-1)=(\mathrm{Sr}^{87})_t-(\mathrm{Sr}^{87})_0, \quad (12)$$

where λ is the decay constant of Rb^{87}, Δt is the elapsed time, and the subscripts r and o stand for "radiogenic" and "t_0." Defining

$$(\mathrm{Sr}^{87})_t/(\mathrm{Sr}^{86})\equiv x \quad (\mathrm{Sr}^{87})_0/(\mathrm{Sr}^{86})\equiv x_0$$
$$(\mathrm{Rb}^{87})_t/(\mathrm{Sr}^{86})\equiv R$$

and substituting in Eq. (12), we obtain

$$x-x_0=R(e^{\lambda\Delta t}-1). \quad (13)$$

In this equation, x, R, and λ are known. Unknown are x_0 and Δt, so that we are dealing with one equation and two unknowns. It has become customary to solve it by combining it with the equation for another meteorite, with a different value of R. Here, the tacit assumption is made that both meteorites have the same age Δt and the same primordial strontium composition x_0. It is also assumed that both meteorites contain Rb of the same isotopic composition, and that they have remained closed systems since t_0. Using subscripts 1 and 2 for the two meteorites, we then obtain

$$x_0=(x_2R_1-x_1R_2)/(R_1-R_2) \quad (14)$$

and

$$\lambda\Delta t=\ln[1+(x_1-x_0)/R_1]. \quad (15)$$

---

[3] The discussion in the early part of this section follows that of Schumacher (1956) in many respects.

Table VI. Isotopic composition of strontium in meteorites.

Meteorite	Class	Sr (ppm)	Rb (ppm)	$\frac{Sr^{87}}{Sr^{86}}$	Reference
Beardsley II	Cg	10 4	14.25	0.960_1	a
Forest City	Ccb	10.2	2.71	0.755_4	a
		11.9_7	3.91	0.754	b
		9.8	3.5	0.756	c
Holbrook	Cik	12.8	2.22	0.739_1	a
Homestead	Cgb	10.6	3.6	0.753	c
Modoc	Cwa	10.1	2.96	0.756_9	a
Richardton	Cca	10.1	2.85	0.756_6	a
Bustee	Bu			0.685	b
Moore County	Eu	79.5	0.16	0.701_3	a
Nuevo Laredo	Ho	84.4	0.37	0.702_7	a
Pasamonte	Ho	82.7	0.22	0.701_2	a
				≤ 0.703	c
Pasamonte (White)		94.7	0.50_0	0.689	b
Pasamonte (Grey)		89.5	0.65_8	0.685	b
Sioux County	Ho	68.8	0.23	0.701_5	a

[a] Gast (1960c, 1961).
[b] Schumacher (1956).
[c] Herzog and Pinson (1956).

Table VI lists all available rubidium and strontium measurements for stone meteorites. Ages calculated for various pairs or combinations of meteorites are given in Table VII.

Before attempting a discussion of these data, we must recognize a limitation of this dating method; namely, it dates a process and not necessarily an object. Suppose a body of primordial material A, of a certain Rb/Sr and Sr^{87}/Sr^{86} ratio, differentiates into a high-rubidium fraction B and a low-rubidium fraction C. At some later time, fragments of B and C, as well as some unchanged A, are recovered as meteorites. Clearly, any two of these can be used to calculate an age by means of Eqs. (14) and (15), but this "age" does not have the same meaning in all cases. For the pair $B-C$, it dates the differentiation process that established the Rb/Sr ratios in these two meteorites. This process represents a physical event in the history of these meteorites, and the date of the process may, therefore, be interpreted as an age. The same numerical value is found, of course, for the two pairs $A-B$ and $A-C$, but here the age applies only to the differentiated member of the pair, B or C. The undifferentiated member A was not affected by the differentiation process; hence, the date of this process has no physical meaning for it. In fact, A could have aggregated as early as 20 Gyr ago, or melted as late as yesterday, yet it would still give the same "age" in Eqs. (14) and (15).

It is clear that no meaningful Rb–Sr age can be assigned to undifferentiated material. The problem is, to recognize such material among the meteorites dated in Table VII. It is also necessary to examine the premises on which Eqs. (14) and (15) were based: contemporaneity of all meteorites and identity of their primordial Sr^{87}/Sr^{86} ratios.

Both problems can be discussed most conveniently in terms of a graphical representation of the data. We rearrange Eq. (12) to the form

$$\frac{(Sr^{87})}{(Sr^{86})_t}=\frac{(Rb^{87})}{(Sr^{86})_t}(e^{\lambda\Delta t}-1)+\frac{(Sr^{87})}{(Sr^{86})_0}. \tag{12a}$$

If Δt is constant, this is the equation of a straight line. On a plot of $(Sr^{87}/Sr^{86})_t$ vs $(Rb^{87}/Sr^{86})_t$, all meteorites of the same age Δt and same initial Sr^{87}/Sr^{86} ratio should lie on a straight line of slope $(e^{\lambda\Delta t}-1)$, an "isochrone." Such a plot of the data in Table VI is shown in Fig. 6.

There appear to be systematic differences between the data of different investigators. The three independently determined Sr^{87}/Sr^{86} ratios of Forest City agree reasonably well with each other (and, for that matter, with those of all other chondrites except Beardsley). However, Schumacher's values for Pasamonte disagree with those of Herzog and Pinson, and of Gast. Gast's figure for Pasamonte also checks with those for two other howardites, Nuevo Laredo and Sioux County, and a eucrite, Moore County.

The rubidium measurements also tend to disagree. Gast's values are the lowest and are probably least affected by contamination. They also check with the neutron-activation results of Webster, Morgan, and Smales (1958) and should, therefore, be given greatest weight.

Gast's points can be represented by an isochrone corresponding to $\Delta t=4.37$ Gyr. Most of the deviant points (with the possible exception of Beardsley) could be shifted on to the line by a small change in the Rb^{87}/Sr^{86} ratio. Thus, it seems that the Rb-Sr fractionations dated by this method occurred during a relatively brief era some 4.37 Gyr ago.

For the chondrites,[4] however, an ambiguity exists. They have a Rb/Sr ratio intermediate between that of Beardsley and the Ca-rich achondrites. Qualitatively, they, therefore, occupy a position comparable to the undifferentiated material A in our foregoing example. In fact, chondrites have long been regarded as average samples of nonvolatile planetary matter

Table VII. $Rb^{87}-Sr^{87}$ ages of meteorites[a]

Meteorite 1	Meteorite 2	Age (Gyr)	Estimated error %	Reference
Pasamonte	Forest City	4.54	±5	b
Av. 4 achondrites	Forest City	4.6		c
Av. 4 achondrites	Modoc	4.2		c
Av. 4 achondrites	Richardton	4.20		c
Av. 4 achondrites	Beardsley	4.4		c
Av. 4 chondrites	Beardsley	4.16		c

[a] Half-life of Rb^{87} –46Gyr (Flynn and Glendenin 1959).
[b] Schumacher (1956).
[c] Gast (1960c, 1961).

---

[4] The term "chondrites" in this discussion usually means "chondrites exclusive of Beardsley."

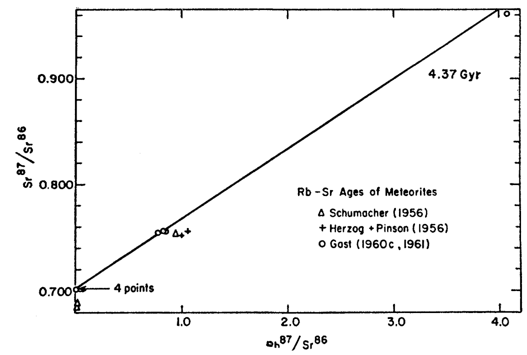

FIG. 6. $Rb^{87}-Sr^{87}$ ages of stone meteorites. The points at $Rb^{87}/Sr^{86}\approx 0$ represent howardites and eucrites; the point at ~ 4, Beardsley; the points at ~ 0.8, the remaining chondrites. A least-squares fit of Gast's points gives an isochrone with a slope corresponding to $t=4.37$ Gyr (with a Rb^{87} half-life of 46 Gyr; Flynn and Glendenin 1959).

(Noddack and Noddack 1934, Urey 1952). Without a standard of reference, we have no way of deciding whether they ever underwent a Rb/Sr fractionation. Hence, we may not rightly assign the Rb-Sr ages in Table VII to the chondrites, although at least one chondrite, Beardsley, became enriched in Rb around 4 to 4.5 Gyr ago, and the other chondrites appear to have suffered extensive processing at that time.

The nature of the Rb-Sr-fractionation process dated is not quite certain. The enrichment in strontium of the achondrites is undoubtedly due to igneous differentiation, and the depletion in Rb may, in principle, be due to the same cause. It is true that, under terrestrial conditions, Rb normally follows K, since the similarity of ionic radii ($K^+=1.33$ A; $Rb^+=1.47$ A) permits isomorphous substitution in potassium minerals. But, in meteorites, potassium minerals are unknown, and the larger size of Rb^+ may well act to exclude it from minerals in which the smaller K^+ still has an appreciable solubility. In fact, Taylor and Heier (1958) found that certain Ca-rich feldspars tend to discriminate strongly against Rb.

Urey (1957b) and Gast (1960a) have argued, however, that the depletion in rubidium is due to selective volatilization. Their view is supported by the discovery that much of the "excess" K, Rb, and Cs in Beardsley is water-soluble (Gast 1960b, 1961), as would be expected for alkalis condensed from the vapor phase. It, therefore, seems likely that fractionation of alkalis by volatilization occurred in at least one instance. Whether the same process was also responsible for the alkali depletion of the calcium-rich achondrites is not yet certain. In any case, the fractionation of Rb and Sr requires high temperatures, above the melting point of silicates, and the Rb-Sr method may, therefore, be taken to date the end of the last melting era that fractionated these two elements. To a good approximation, this event should coincide with the solidification of the molten regions in the meteorite parent bodies.

4.2 Rhenium-187-Osmium-187 Ages

This method is based on a pure β decay and is, therefore, quite similar in principle to the Rb^{87}-Sr^{87} method. In practice, however, there are certain limitations that have restricted its usefulness.

The most serious handicap faced by this method is the lack of geochemical fractionation between parent and daughter element. Both Re and Os are believed to be stongly siderophile (Brown and Goldberg 1949, Goldschmidt 1954). Their chalcophile tendencies are not well known, but the only modern data available indicate that the Os^{187}/Os^{186} ratios and, hence, the Re–Os ratios are virtually identical in Odessa troilite and metal phase (Herr, Hoffmeister, Langhoff, Geiss, Hirt, and Houtermans 1960).

Another difficulty is the uncertainty in the half-life of Re^{187}. A direct counter measurement is exceedingly difficult owing to the low decay energy (<8 kev).[5] The best estimates are, therefore, obtained from Re and Os measurements on minerals of known age. The original estimate of 62 (+6, −7) Gyr (Herr and Merz 1958) has recently been revised to 43±5 Gyr (Herr, Hirt, and Hoffmeister 1961).

Table VIII gives Re^{187}/Os^{186} and Os^{187}/Os^{186} ratios for fourteen iron meteorites analyzed by Herr *et al.* (1960) and by Herr, Hoffmeister, Hirt, Geiss, and Houtermans (1961). The same data are also shown in Fig. 7.

Clearly, the errors are much larger in this case than in the Rb^{87}-Sr^{87} method. It is, therefore, hardly worthwhile to calculate ages for individual meteorite pairs from Eqs. (14) and (15). Instead, it may be best to assume on the strength of the Rb^{87}-Sr^{87} and Pb^{207}/Pb^{206} results that the meteorites are congenetic within the accuracy of the data and that they initially contained primordial osmium of identical isotopic composition. Then, the slope of the best straight line drawn through the points in Fig. 7 should give the age, and the intercept,

TABLE VIII. Rhenium and osmium analyses of iron meteorites (Herr *et al.* 1960; Herr, Hoffmeister, Hirt, Geiss, and Houtermans 1961).

Meteorite	Os^{187}/Os^{186}	Re^{187}/Os^{186}
Bethany	1.094±0.008	3.50±0.23
Canyon Diablo	1.127±0.019	5.02±0.24
Carbo	1.056±0.011	3.36±0.26
Casas Grandes	1.128±0.005	5.77±0.97
Gibeon	1.110±0.012	3.88±0.48
Henbury	1.073±0.011	3.96±0.44
Linwood	1.016±0.014	3.26±0.41
Negrillos	1.015±0.023	3.77±0.40
Odessa	1.086±0.010	3.99±0.19
Piñon	1.015±0.019	2.55±0.26
Tlacotepec	1.004±0.018	2.98±0.19
Tocopilla	1.409±0.012	8.10±0.99
Toluca	1.063±0.013	3.55±0.50
Treysa	1.20 ±0.11	6.62±1.10

---

[5] This low decay energy may also lead to a slight dependence of half-life on chemical state (Herr *et al.* 1960).

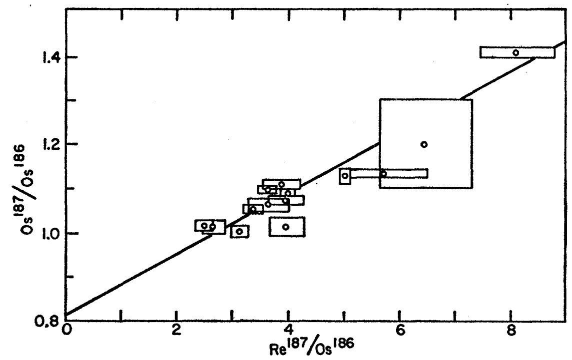

FIG. 7. $Re^{187}-Os^{187}$ ages of iron meteorites. (Herr, Hoffmeister, Hirt, Geiss, and Houtermans 1961). The straight line represents a least-squares fit of the data. Its slope gives an age of 4.0±0.8 Gyr for $t_{\frac{1}{2}}=43$ Gyr.

the primordial Os^{187}/Os^{186} ratio (Herr, Hoffmeister, Hirt, Geiss, and Houtermans 1961).

The constants thus obtained by a weighted least-squares fit are

$$\lambda t = 0.063 \pm 20\% \quad (t = 4.0 \pm 0.8 \text{ Gyr for } t_{\frac{1}{2}} = 43 \text{ Gyr})$$
$$(Os^{187}/Os^{186})_0 = 0.83.$$

What then is the physical meaning of this age? Evidently it dates a Re-Os fractionation. But, it is not immediately obvious whether this fractionation took place among the iron meteorites themselves, or whether it involved another phase not represented in the present measurements.

The former case is attractive because of its simplicity; certainly the division of the data in Fig. 7 into three clusters is suggestive of the differentiation of *A* into *B* and *C*, as discussed in Sec. 4.1. In that case, the usual ambiguity exists: The age applies only to the differentiated fractions, not to the primordial material *A*, or any material derived from it that has kept the same Re/Os ratio. Again, we have no safe criterion for identifying such material, though the closeness of the Os^{187}/Os^{186} ratio of the middle cluster to the terrestrial ratio (1.070–1.086, Herr *et al.* 1960) makes this cluster the prime suspect.

However, it is rather unlikely that the fractionation took place among the iron meteorites themselves, for this would imply that all iron meteorites originated in the same body. This is not consistent with the division of the iron meteorites into 4 clusters of markedly different Ga and Ge content (Goldberg, Uchiyama, and Brown 1951; Lovering, Nichiporuk, Chodos, and Brown 1957), and particularly with the discrete grouping of cosmic-ray-exposure ages (Sec. 2).

Also, there is very little correlation between Ni content, Re/Os ratio, and the absolute Re and Os contents (Table IX). Negrillos, a hexahedrite, and Dayton, a Ni-rich ataxite, have virtually the same Os/Re ratio though the absolute Re and Os contents of Negrillos are 10^3 times higher. In contrast, another Ni-rich ataxite, Tlacotepec, contains nearly the same amounts of Re and Os as Negrillos, and a slightly higher Re/Os ratio. Tocopilla, another Chilean hexahedrite, has an intermediate Re and Os content, but its Os/Re ratio is the lowest of any of the meteorites studied.

One must, therefore, consider the possibility that the Re-Os fractionation involved at least one other phase. The nature of this phase is still quite uncertain. It is not likely to have been silicate, since the lithophile tendencies of Re and Os are known to be very weak (Herr, Hoffmeister, Hirt, Geiss, and Houtermans 1961). The troilite phase of iron meteorites would seem to offer a slightly more attractive possibility, but in at least one case (Odessa) no significant difference was found in the Os^{187}/Os^{186} ratio and, hence, in the Re/Os ratio of two samples differing materially in troilite content. (Herr *et al.* 1960). Perhaps a more likely prospect is the hypothetical, troilite-phosphide layer postulated by Fish, Goles, and Anders (1960) as the locus of chalcophile elements that are underbundant in chondrites. This layer would be immiscible with metal at temperatures not much above 1800°K, and would most probably be in isotopic exchange with the metal phase until the solidification of the latter (Vogel 1961). The Re-Os method would then date this solidification.

It would be interesting to test this hypothesis by direct measurements on possible representatives of the troilite-phosphide layer, such as Soroti.

The measurements of Herr *et al.* also have interesting implications for the history of the Earth. On the one hand, a comparison of the average crustal and meteoritic Os^{187}/Os^{186} ratios indicates the extent of any Re/Os fractionation during the formation of the Earth's core. On the other hand, the discovery of terrestrial osmiridium samples of very low Os^{187}/Os^{186} ratio[6] offers hope of finding material of even greater age than the oldest known rocks. The high melting point and chemical inertness of osmiridium give it an excellent chance of surviving repeated cycles of magmatic differentiation.

4.3. Lead-207/Lead-206 Ages

This method has two intrinsic advantages over the preceding ones: Only isotopic rather than elemental

TABLE IX. Rhenium and osmium contents of meteorites with extreme nickel contents.[a]

Meteorite	Class	Ni %	Re ppm	Os ppm	Os/Re	Os^{187}/Os^{186}
Dayton	D_I	18.10	0.0050	0.047	9.4	
Tlacotepec	D_I	16.23	2.92	39.6	13.6	1.004±0.018
Negrillos	H	5.32	4.80	50.4	10.5	1.015±0.023
Tocopilla	H	5.43	0.25	1.29	5.12	1.409±0.012

[a] From Herr *et al.* (1960); Herr, Hoffmeister, Hirt, Geiss, and Houtermans (1961).

---

[6] A ratio of 0.882 was observed in a sample from Witwatersrand, South Africa.

TABLE X. Lead-207/lead-206 ages of stone meteorites.

			Lead-isotopic composition				
Meteorite	Class	Pb content (ppm)[a]	$\frac{206}{204}$	$\frac{207}{204}$	$\frac{208}{204}$	Pb^{207}/Pb^{206} age (Gyr)[b]	Reference
Beardsley	Cg	0.13	13.6_7	12.4_0	31.8_5	4.27	c
		0.15					f
Elenovka	Ck (?)	*0.48*	21.54	16.94	39.86	4.44	e
Forest City	Ccb	*0.4*	19.27	15.95	39.05	4.5	d
		0.15					f
		0.09					c
Holbrook-*N*	Cck	*0.28*	21.2_0	15.2_2	35.5_3		c
Holbrook-*M*1			16.6_8	14.5_7	35.6_8	4.54	c
Holbrook-*M*2			16.0_4	14.0_4	33.8_2	4.48	c
Holbrook		*0.38*					f
Indarch	Cek	1.7			26.8±1.2		f
Kunashak	Ck	*0.53*	19.64	16.24	40.04	4.52	e
Mighei	K	1.3			31.1±1.2		f
Modoc	Cwa	*0.9*	19.48	15.76	38.21	4.4	d
		0.06					f
Norton County	Au	*0.57*	22.75	15.87	37.70	4.02	e
Nuevo Laredo	Ho	0.7	50.28	34.86	67.97	4.6	d
		0.76			59±8		f
Orgueil	K	2.3			30.3±1.2		f
Plainview	Ciab	*0.46*	14.8_7	13.6_1	34.0_0	4.58	c
Richardton	Cca	0.05_5	>38.16	>27.70	>56.27	4.61	c
Saratov	Cci	*0.40*	19.53	16.70	40.25	4.65	e

[a] Lead values that are clearly in error are printed in italics.

[b] Calculated with respect to Patterson's average of Canyon Diablo and Henbury lead.

[c] Hess and Marshall (1960).

[d] Patterson (1955).

[e] Starik, Shats, and Sobotovich (1958).

[f] Reed, Kigoshi, and Turkevich (1960).

abundances need to be measured, and the decay constants are known with high accuracy. Unfortunately, these advantages are offset in large measure by severe contamination problems.

The derivation of the method follows. Let

U_5 = number of U^{235} atoms at present time t
U_8 = number of U^{238} atoms at present time t
P_4 = number of Pb^{204} atoms at present time t
P_6 = number of Pb^{206} atoms at present time t
P_7 = number of Pb^{207} atoms at present time t
k = present U^{238}/U^{235} ratio

λ_5, λ_8 = decay constants of U^{235}, U^{238}

superscript ^{0} = at time t_0 of meteorite formation
superscript * = radiogenic

$$\Delta t = t - t_0 = \text{“age” of meteorite.}$$

Then,

$$P_7^* = U_5^0 - U_5 = U_5(e^{\lambda_5 \Delta t} - 1) \tag{16a}$$

$$P_6^* = U_8^0 - U_8 = U_8(e^{\lambda_8 \Delta t} - 1). \tag{16b}$$

Dividing (16a) by (16b), we obtain

$$P_7^* = P_6^* \frac{(e^{\lambda_5 \Delta t} - 1)}{k(e^{\lambda_8 \Delta t} - 1)}. \tag{17}$$

This equation could be used directly to calculate the age, if the proportion of radiogenic lead in the meteorite were known. Unfortunately, the meteorites always contain nonradiogenic lead, and neither its amount not its isotopic composition can be estimated independently. This difficulty is circumvented in the same manner as in the case of Rb-Sr ages, by combining the data for two meteorites. It is again assumed that both meteorites initially contained lead and uranium of the same isotopic composition, and that they have the same age.

Normalizing the data to Pb^{204} (which is not produced by radioactive decay), writing two separate equations (17) for meteorites A and B, and subtracting them from one another, we obtain:

$$\frac{(P_7/P_4)_a - (P_7/P_4)_b}{(P_6/P_4)_a - (P_6/P_4)_b} = \frac{(e^{\lambda_5 \Delta t} - 1)}{k(e^{\lambda_8 \Delta t} - 1)}. \tag{18}$$

This equation can be solved for Δt by graphical methods. To minimize error, one always chooses meteorite pairs that differ greatly in their U/Pb ratio and, hence, in the isotopic composition of their lead. The U/Pb ratio is particularly low in troilite from iron meteorites, so low, in fact, that the isotopic composition of this lead is virtually primordial, not having been perceptibly altered by radioactive decay of U and Th during the last 4.6 Gyr. Because of its extreme isotopic composition, "primordial" lead[7] is commonly used as "meteorite B" in age calculations. However, any age thus calculated is still afflicted with the usual ambiguity: Does it refer to meteorite A, meteorite B, or both?

---

[7] That is, the average of Canyon Diablo and Henbury troilite leads (Patterson 1955, 1956). More recently, Murthy and Patterson (1961) suggested a revised set of values based on a larger number of measurements from different laboratories ($P_6/P_4 = 9.56$; $P_7/P_4 = 10.42$; $P_8/P_4 = 29.71$).

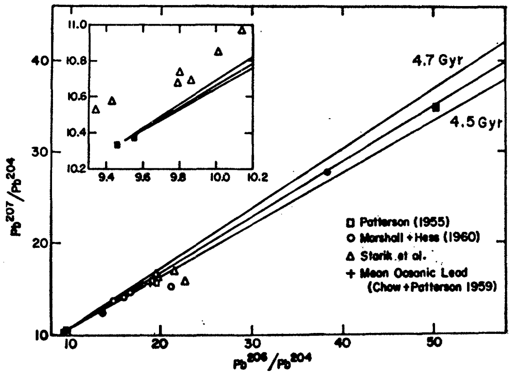

FIG. 8. $Pb^{207}-Pb^{206}$ ages of meteorites. Solid points represent meteorites in which measured lead and uranium contents are consistent with each other. Insert shows "primordial" lead data. Note systematically higher Pb^{207}/Pb^{204} ratios of Starik's points.

Table X summarizes meteoritic lead-isotope data and the ages calculated therefrom.

Just as in the case of the Rb-Sr and Re-Os ages, it is again more instructive to analyze the data in terms of an isochrone. Equation (17) can be written in the form

$$\frac{P_7}{P_4}=\frac{P_6}{P_4}\left[\frac{(e^{\lambda_5\Delta t}-1)}{k(e^{\lambda_8\Delta t}-1)}\right]-\frac{P_6^0}{P_4}\left[\frac{(e^{\lambda_5\Delta t}-1)}{k(e^{\lambda_8\Delta t}-1)}\right]+\frac{P_7^0}{P_4}. \qquad (17a)$$

For a constant Δt and constant primordial lead composition, this is obviously the equation of a straight line. On a plot of Pb^{207}/Pb^{204} vs Pb^{206}/Pb^{204}, all meteorites of identical primordial-lead composition should lie on a single isochrone. As seen in Fig. 8 most meteorites do indeed fall within 0.1 Gyr of the 4.6 Gyr isochrone.

Unfortunately, most of these ages are not nearly as well-determined as the close fit to the isochrone would seem to indicate. In most cases, the measured uranium and thorium concentrations in chondrites, as determined by neutron-activation analysis, are too low by factors of 2–10 to account for the apparent content of radiogenic lead (Hamaguchi, Reed, and Turkevich 1957). Both lead and uranium are strongly enriched in the Earth's crust relative to the meteorites, and the problem of contamination with terrestrial lead is, therefore, exceedingly severe. Only in three cases (Beardsley, Nuevo Laredo, and Richardton) are the lead and uranium data in tolerable agreement.

In some cases, the contamination problem may be of less consequence than is indicated by the lead-abundance data. The lead for the isotopic-composition measurement is usually separated by direct sublimation while the concentration measurement is done on a separate and often smaller sample, with much more extensive chemical processing. Unfortunately, modern terrestrial lead (e.g., the point for mean oceanic lead) also fits the 4.6 Gyr isochrone, since the Earth seems to have fractionated its Pb and U at about the same time as the meteorites (Patterson 1956, Chow and Patterson 1959). Hence, the mere agreement of a given lead datum with the isochrone tells nothing about the degree of contamination and very little about the age of the meteorite, unless there is independent evidence on this point.

Taken at face value, the two stray points in Fig. 8 would seem to indicate appreciable age differences among the meteorites. However, terrestrial leads exist that do not fit the isochrone ("*B*"- and "*J*"- type leads, Eberhardt, Geiss, and Houtermans 1955). Contamination with leads of this type may well account for the unusual isotopic composition reported for Holbrook-*N* (a sample with a 25-yr exposure to groundwater, in contrast to relatively unaltered Holbrook-*M*) and Norton County.

A curious problem is presented by the iron meteorites (Table XI). Two samples of Toluca metal and troilite gave lead of approximately primordial isotopic composition, whereas two other troilite samples from the same meteorite gave much more radiogenic lead. Similar discrepancies were found for Bischtübe and Henbury (Patterson 1955, Starik *et al.* 1959, 1960). For eight other meteorites, the latter authors found only radiogenic lead.

In Sikhote-Alin, the measured amount of uranium is too low by a factor of 100 to account for growth of the radiogenic lead in 5 Gyr (Fireman and Fisher 1961). In Toluca, too, it is quite certain that the troilite contains far too little uranium to account for the observed amounts of radiogenic lead. Uranium contents obtained by three different methods are <13 ppb (Reed *et al.* 1960), <10 ppb (Murthy 1961a), and 10 ppb (Goles and Anders 1961c). The isotopic composition of the radiogenic Toluca lead falls between the 4.6- and 4.7-Gyr isochrones and, thus, fits rather well into the meteorite array. The same is true of the radiogenic lead from other iron meteorites (Starik *et al.* 1959, 1960), though all these values show a systematic tendency to lie slightly *above* the 4.6-Gyr isochrone.

In the case of well-determined terrestrial lead data, such an apparent excess of Pb^{207} is usually attributed to a Pb-U separation at some previous time and can, in fact, be used to calculate the date of such a separation. Murthy (1961a), therefore, postulated a preterrestrial event that added radiogenic lead from the silicate phase of the meteorite parent bodies to parts of the iron meteorites some 0.5 Gyr ago. Marshall and Hess (1961) had previously mentioned this possibility. Actually, this conclusion does not seem justified by the data, since Starik's primordial-lead points, too, are high in Pb^{207} relative to Patterson's values (see insert, Fig. 8). This discrepancy implies either systematic errors of measurement from one laboratory to another, or the existence of more than one type of primordial lead. In neither case would it be justified to explain these slight departures from the isochrone in terms of recent preterrestrial events.

It seems more likely that the radiogenic lead was introduced into the troilite by an ion-exchange mechanism during *terrestrial weathering*. The solubility product of PbS ($\sim 10^{-28}$) is much smaller than that of FeS ($\sim 10^{-20}$) so that troilite in contact with ground water containing the usual amounts of Pb^{++} will gradually become enriched in lead. It is certainly striking that the "radiogenic" Toluca troilite has a tenfold higher lead content than the "primordial" sample. Also, Starik *et al.* (1960) report a fivefold decrease in the lead content from the surface to the interior of the Santa Catharina meteorite, and a similar decrease in Sikhote-Alin and Chinge. All but one of the irons with radiogenic lead are finds, and the exception, Sikhote-Alin, is reported to have such a low lead content (0.03 ppm in metal; 1.0 ppm in troilite) that the risk of contamination during chemical processing would appear to be particularly great.

The isochrone is thus defined chiefly by the iron meteorites with primordial lead and by the three stones for which the lead data are consistent with the uranium content. Allowing for experimental error, the agreement with the 4.6-Gyr isochrone is fairly satisfactory, and it thus seems that the premises of the Pb-Pb method, as formulated by Patterson in 1955, are still valid. Nonetheless, many more data will be required to establish this point with certainty.

The event dated by the Pb-Pb method is the separation of lead from uranium and thorium. In the case of the irons, this event probably coincided with the separation of the metal and silicate phases in the meteorite parent bodies. The howardite Nuevo Laredo, on the other hand, seems to have undergone a threefold differentiation with respect to primordial matter of the approximate composition of carbonaceous chondrites: it lost its Ni and other siderophile elements; it (or, more precisely, its silicate phase) became enriched in Ca, Al, U, and Th, while being depleted in Mg, Na, and the heavier alkalis; and it lost its chalcophile elements, such as Tl, Pb, and Bi (Reed *et al.* 1960). For convenience, these elements are called chalcophile, although Vogel (1961) has pointed out that their concentration in the troilite phase is not an indication of a high affinity for sulphur, but of the immiscibility of these metals with iron. Within the present resolving power of the Pb-Pb method, the last two separations seem to have occurred simultaneously. This tends to confirm the impression that the era of meteorite synthesis was short.

Taken as a group, the chondrites have a U/Pb ratio intermediate between those of the irons and the Ca-rich achondrites. In the Rb/Sr and Re/Os methods, it proved difficult to date such "intermediate" material, since there was no assurance that it had ever undergone the differentiation process in question. In the present case, the situation is more fortunate. Uranium and thorium do not seem to have suffered any appreciable fractionation from each other or from silicon, judging from both the constancy of the chondrite analyses (Hamaguchi *et al.* 1957; Bate *et al.* 1959) and their compatibility with the heat-balance data for the Earth, the Moon, and the terrestrial plantets (Urey 1956a, MacDonald 1959). On the other hand, there is good evidence for a fractionation of lead: the ratios of uranium to lead-204 differ markedly from chondrite to chondrite (Reed *et al.* 1960). Although a small, residual ambiguity remains (in a set of n different chondrite leads, only $n-1$ can be safely regarded as differentiated and, hence, datable), it is certain that the lead depletion of the chondrites can be dated by the Pb-Pb method. According to the present data, based entirely on Richardton, this depletion seems to have taken place about 4.6 Gyr ago. Thus, all three methods involving fractionations at the melting temperatures of metal and silicates seem to give virtually the same result: The era of intense melting in the meteorite parent bodies ended about 4.4 to 4.6 Gyr ago.

5. "GAS-RETENTION" AGES

The meteorites contain four long-lived radionuclides that produce noble gases in their decay:

$$K^{40} \xrightarrow[\text{1.25 Gyr}]{\text{electron capture (11\%)}} Ar^{40}$$

$$U^{238} \xrightarrow[\text{4.51 Gyr}]{\alpha,\ \beta^-} Pb^{206} + 8\ He^4$$

$$U^{235} \xrightarrow[\text{0.71 Gyr}]{\alpha,\ \beta^-} Pb^{207} + 7\ He^4$$

$$Th^{232} \xrightarrow[\text{13.9 Gyr}]{\alpha,\ \beta^-} Pb^{208} + 6\ He^4.$$

At high temperatures, these gases diffuse rapidly through silicate lattices. Only at low temperatures can they be accumulated and retained by their parent minerals. These radioactive-decay chains thus offer, in principle, a means for determining the onset of gas retention, or the date of the last heating of the meteorite. For meteorites that have remained cold throughout their later history, this date coincides with the cooling time of the meteorite parent bodies. If this condition is not met, the interpretation becomes more complicated, as is shown in Sec. 5.3.

5.1 K-Ar Ages

The K-Ar age t, can be calculated from the Ar^{40} and K^{40} content of the meteorite by the equation

$$t = \frac{1}{\lambda} \ln \left[1 + \frac{Ar^{40}}{K^{40}} \frac{(1+R)}{R} \right], \qquad (19)$$

TABLE XI. Lead isotopic composition in iron meteorites.

			Lead isotopic composition			
Meteorite	Class	Pb content (ppm)	$\frac{206}{204}$	$\frac{207}{204}$	$\frac{208}{204}$	Reference
Aroos	Om	0.17	10.14	10.97	30.18	b
Aroos troilite		2.3	10.01	10.85	30.78	b
Bischtübe	Og	0.18	9.80	10.74	30.08	b
Bischtübe "troilite"[a]		7.5	17.72	15.74	38.40	b
Burgavli	O	0.24	9.34	10.53	30.28	b
Burgavli troilite		7.67	9.79	10.68	30.27	b
Canyon Diablo	Og	0.13	9.43	10.58	29.80	c
Canyon Diablo troilite		18	9.46	10.34	29.44	d
		4.9			28.2±1.0	e
Henbury	Om	0.13	18.13	15.96	38.70	e
Henbury troilite		4.9	18.41	15.78	39.00	c
Henbury troilite		5	9.55	10.38	29.54	d
Toluca	Om	0.16	9.87	10.70	30.36	b
Toluca troilite		5.4			29±3	e
Toluca troilite		59	17.18	15.48	37.96	f
Toluca troilite		49.6	16.87	15.03	37.12	g

[a] The authors suggest that this inclusion might be some sulfide mineral other than troilite, possibly much richer in U and Th.
[b] Starik, Sobotovich, Lovtsyus, Shats, and Lovtsyus (1960).
[c] Starik, Sobotovich, Lovtsyus, Shats, and Lovtsyus (1959).
[d] Patterson (1955).
[e] Reed, Kigoshi, and Turkevich (1960).
[f] Marshall and Hess (1961).
[g] Murthy (1961a).

where λ is the (total) decay constant of K^{40} and R is the "branching" ratio, i.e., the ratio of electron capture to β^- emission events in the decay. The presently accepted value of R is 0.124, but, until recently, values ranging from 0.064 to 0.136 were used.

The first meteorite-age determinations by this method were made by Gerling and Pavlova (1951). Since then, many additional data have become available (Tables XII and XIII).

Not all of the reported values are of equal reliability. The potassium content of meterorites is quite low, and conventional, wet-chemical methods, therefore, tend to give high results. Special techniques must be used in the isolation of potassium (Wasserburg and Hayden 1955a; Edwards and Urey 1955; Geiss and Hess 1958), and the determination itself is best done by instrumental methods, such as isotope dilution or flame photometry. Edwards (1955) has shown that the mean potassium content of chondrites (except for carbonaceous chondrites) is ~850 ppm, with very little dispersion about the mean. The departure of the individual potassium contents in Table XII from this mean, therefore, provides a rough index of the reliability of the data. Only in the case of Beardsley and the carbonaceous chondrites is a large deviation from the mean well substantiated.

The argon measurements, too, can be in error. Atmospheric contamination was a problem in many of the early experiments, but has since been brought under control. A more serious source of error is the incomplete extraction of argon. Experiments by Geiss and Hess (1958) show that quite drastic conditions (e.g., heating for 6 hr at 1200°C in a borax flux) are required to liberate all gases. Mere heating to 1200°C in the absence of a flux, as practiced by Gerling and Rik (1955) and by Trofimov and Rik (1955), gave incomplete argon yields. This was demonstrated specifically in the case of Bjurböle, where the ages determined by the two techniques differ by 0.7 Gyr (Table XII).

If all errors are carefully minimized, the reproducibility of the method is excellent (note, for example, Beardsley, Forest City, and Pesyanoe, each of which was studied by two independent groups of investigators). On the other hand, many of the earlier results are likely to be low, owing to potassium contamination and incomplete argon extraction. Such doubtful values have been italicized in Table XII.

An ingenious method that permits determination of both the potassium and argon content in the same sample was developed by Wänke and König (1959). When a meteorite is irradiated in a neutron flux containing an appreciable proportion of fast neutrons, three radioactive isotopes of argon are produced:

$$Ca^{40}(n,\alpha)Ar^{37}(\text{electron capture, 35 days})$$
$$K^{39}(n,p)Ar^{39}(\beta^-,\ 325\text{ yr})$$
$$Ar^{40}(n,\gamma)Ar^{41}(\beta^-,\ 1.82\text{ hr}).$$

The last two activities can serve as measures of the potassium and Ar^{40} contents of the meteorite.

A particular advantage of this method is the fact that both the potassium and the argon-40 give rise to *argon* activities. Only the ratio Ar^{40}/K^{40} is needed for an age determination, and it thus suffices to measure the *ratio* Ar^{41}/Ar^{39}, rather than the absolute amounts of both activities. Since both potassium and argon-40 are likely to reside in the same phase (unless reheating has caused redistribution of the argon-40 in the meteorite), quantitative argon extraction and recovery is not required. Unfortunately, Ar^{36} and Ar^{38} cannot be measured by this method, so that the amount of atmospheric, cosmogenic, or primordial argon cannot be determined.

Stoenner and Zähringer (1958) have also attempted to measure K-Ar ages of *iron* meteorites by activiation analysis, using 12.4-hr K^{42}, produced by (n,γ) reaction on K^{41}, as an index of the potassium content. Assuming normal isotopic composition of the potassium, they obtained ages ranging from 5 to 13 Gyr, very much greater than the general run of meteorite ages. They were careful to exclude all obvious sources of error, and, although these extraordinary results were met with some skepticism, no outright refutation of this work has appeared in print.

However, a rather suggestive trend is present in their data (Stoenner and Zähringer 1958, Tables 1 and 2). With one exception, the high Ar^{40}/K ratios (and the high ages) are associated with the lowest K contents. It must be remembered, of course, that the K^{40}-Ar^{40} age depends on the K^{40} content, which Stoenner and Zähringer did not measure directly, but calculated from the measured K^{41} content assuming normal isotopic composition. But, as shown by Voshage and Hintenberger (1959, 1961) and by Honda (1959), appreciable amounts of K^{40} are produced in the spallation of iron by cosmic rays, the K^{40} content rising from its normal value of $1.18\times10^{-2}\%$ to 5% or more. Hence, it is essential to determine the extent of a correction for cosmogenic K^{40} and for directly produced Ar^{40} as well.

Stoenner and Zähringer assumed $He^3/Ar^{40}=170$, but recent data for the He^3/Ar^{38} ratio (Signer and Nier 1960; Fisher and Schaeffer 1960; Signer and Nier 1961) combined with the calculated cross-section ratio Ar^{40}/Ar^{38} (Goel 1960) suggested that a value in the neighborhood of 50 might have been more appropriate. This would reduce to zero the "radiogenic" Ar^{40} component in meteorites with a high content of cosmogenic gases (Stoenner and Zähringer 1958, Table 1).

In the other meteorites, the correction for cosmogenic Ar^{40} would be smaller, but it is not unlikely that at least part of the remaining Ar^{40} is due to atmospheric contamination.

Also, the content of cosmogenic K^{40} in the Aroos meteorite has recently been measured (0.51 ppb, Stauffer and Honda 1961). Using this value and the He^3 content of Aroos measured by Signer and Nier (1961), 655×10^{-8} cc (at STP), to compute a correction for cosmogenic K^{40} in the meteorites dated by Stoenner and Zähringer, one finds that the true K^{40} content was systematically underestimated by factors of up to 1000. With a single exception, substantial corrections in the K^{40} content are required even for those meteorites whose He^3 content is low (Canyon Diablo and Toluca).

TABLE XII. "Gas-retention" ages of chondrites.[a]

Meteorite	Class	K[b] ppm	$Ar^{40}\times10^{-6}$ cc/g (at STP)	K−Ar age Gyr	U−He age Gyr	References
Abee	Cek				*3.6*	d
Akaba	Cw	1000	45.0	3.57	3.4	e,f,g
		1040	62.1	3.8	3.2	h, i
Alfianello	Ci				*0.84*	f
Beardsley	Cg	1010	71.3	4.3	3.6	j, g
		1000	71.0	4.3	3.8	k, l
Beddgelert	Cs	770	20.2	2.78	0.5	e, g
Benton	C	[850]	44.5	3.8	3.6	m
Bjurböle	Cc	1000	49.3	*3.6*	4.0	n, f
		840	60.5	4.32	4.2	k, l
Breitscheid	Cg	820	32.0	3.33	1.4	e,o,p,g
Bruderheim	C			1.6	1.5	q
Colby, Kansas	Cck	[870]	41.1	3.7		z
De Nova	Ccwka	[870]	39.6	3.65		z
Elenovka	Ck(?)	1000	57.2	4.0	4.0	n, f
		930	62.6	4.19	3.8[c]	r
					4.1	s
Felix	K	420	32.8	4.5		t
Forest City	Ccb	831	52.4	4.15		j, k
		830	53.6	4.15		u
Glasatovo	Cga	1300	45.7	*3.1*		n
Holbrook	Cck	880	66.2	4.4	4.4	k, l
		880	59.8	4.2	*0.9*	bb,f
Hugoton	Csb	790	51.0	4.6		z
Ivuna	K	750	6.4	1.4		t
Kainsaz	Cs	500	27.0	*3.9*		n
Kandahar	C	[850]	62.0	4.3	3.4	m
Khmelevka	Ck	[870]	6.3	1.3		z
Krymka	C	970	52.8	3.84	*5.2*[c]	r
Kunashak (gray)	Ckab	650	2.4	0.7	0.55	n, f
Kunashak (black)	Ckab	900	2.7	1.2		n
La Lande	Cck	[870]	9.3	1.65		z
Lancé	K	>380	20.5	<3.9		t
Manbhoom	CAm	860	56.0	4.2		z
Marion	Cwa	870	54.0	4.08		i
Mezel	Cia	[850]	27.5	3.1	1.3	m
Mighei	K	400	28.7	4.3		n
Mocs	Cwa	870	62.0	4.30	2.4	k, g
Modoc	Cwa	830	55.0	4.18	3.0	e, g
Mokoia	K	380	14.9	3.4		t
Monte das Fortes	C				4.0	s
Monze	Cw	1190	≤16.3	≤1.9		i
Murray	K	380	5.0	1.9		t
		260	<6.53	<2.77		v
Murray	K			2.51		aa
New Concord	Cia				1.0	l
Ochansk	Ccb	650	52.1	4.5	4.0	n
Pervomaiskii (gray)	Ckia	800	2.5	0.65	0.63	n, f
Pervomaiskii (black)	Ckia	1250	15.2	1.8	0.94	n, f
Pultusk	Cga	770	39.6	3.74	1.8	e, g
Ramsdorf	Cb	[850]	1.6	0.37	0.4	w
Richardton	Cca	830	54.0	4.15	3.8	k, l
		730	53.2	4.47		v
Richardton	Cca	[870]	51.5	4.1		z
Richardton	Cca			4.59		aa
Rochester	Cc	[870]	61.4	4.4		z
St. Michel	Cw	910	54.0	4.00	1.9	k, l
Saratov	Ccg	1050	54.2	3.7	3.8	n, f
		1700	53.3	*3.0*	<0.015	x, y
Sevrukovo	Cs	900	17.0	2.3	<0.04	n, y
Temple	C				2.8	g
Zhovtnevyi	Cia	1800	57.2	*3.0*	4.2	n, f, x
					0.03	y
Zvonkovoye	Ck	990	60.8	4.04	4.1[c]	r

[a] Ages that are likely to be in error are printed in italics.
[b] Estimated values are enclosed in brackets.
[c] These ages were recalculated from the total helium content given by the authors, assuming a uranium content of 11 ppb and a cosmogenic He^4 content of 1×10^{-6} cc/g (at STP).
[d] Begemann, Eberhardt, and Hess (1959).
[e] Wänke and König (1959).
[f] Reed and Turkevich (1957).
[g] Hintenberger, König, and Wänke (1961).
[h] Reasbeck and Mayne (1955).
[i] Thomson and Mayne (1955).
[j] Wasserburg and Hayden (1955a).
[k] Geiss and Hess (1958).
[l] Eberhardt and Hess (1960).
[m] Geiss, Oeschger, and Signer (1960).
[n] Gerling and Rik (1955).
[o] König, Wänke, and Mayne (1959).
[p] Ebert, Hernegger, König, and Wänke (1959).
[q] Signer (1961a).
[r] Burkser, Kotlovskaya, and Zaydis (1958).
[s] Geiss, Hirt, and Oeschger (1960).
[t] Stauffer (1961a).
[u] Folinsbee, Lipson, and Reynolds (1956).
[v] Reynolds (1960b).
[w] Goebel, Schmidlin, and Zähringer (1959).
[x] Gerling and Pavlova (1951).
[y] Cherdyntsev and Abdulgafarov (1956).
[z] Stauffer (1961c).
[aa] Krummenacher (1961).
[bb] Wasserburg, Hayden, and Jensen (1956).

TABLE XIII. "Gas-retention" ages of achondrites and stony irons.[a]

Meteorite	Class	K ppm	Ar^{40} 10^{-6} cc (at STP)	K−Ar age Gyr	U−He age Gyr	References
Chervony Kut	Eu	470	9.93	2.7		b
Frankfort	Ho	130	5.1	3.34		c
Johnstown	Di				4.2	m
Juvinas	Eu				3.1	m
Moore County	Eu	210	7.6	3.23		c
Norton County	Au	70	5.3	4.4	≥2.4	e
		230	4.0	*2.3*		b
Nuevo Laredo	Ho	440	12.9–17.7	*3.1–3.6*	0.45	l, f
Padvarninkai	Sh	1000	5.8	1.0		g
Pasamonte	Ho	430	22.5	3.80	3.6	c, e
		340	23.6	4.25	3.6	h, m
Pesyanoe	Au		46.1	4.2		j
			45.9			i
Shergotty	Sh	1500	4.0	0.56		c
Sioux County	Ho				3.0	m
Stannern	Eu	570	16.8	2.95		b
				3.6	3.6	m
Yurtuk	Am	430	9.8	2.55		b
Brenham	P	190	7.8	*3.5*		n
		<15	1.0			c

[a] Ages that are likely to be in error are printed in italics.
[b] Vinogradov, Zadorozhnyi, and Knorre, (1958).
[c] Geiss and Hess (1958).
[e] Eberhardt and Hess (1960).
[f] Reed and Turkevich (1957).
[g] Gerling and Rik (1955).
[h] Wänke and König (1959).
[i] Stauffer (1961a).
[j] Gerling and Levskii (1956).
[k] König, Wänke, and Mayne (1959).
[l] Reynolds and Lipson (1957).
[m] Hintenberger, König, and Wänke (1961).
[n] Thomson and Mayne (1955).

If the cosmic-ray irradiation of the meteorite began abruptly, at a time given by the exposure age, only a fraction of the cosmogenic K^{40} would have decayed, so that the correction to the K-Ar age will be small. However, the correction would be larger, if the onset of the cosmic-ray irradiation had been gradual, such as in the case of multiple breakups.

Another possibility that must be considered, in the case of Canyon Diablo at least, is preferential loss of He^3. It is known that many specimens of Canyon Diablo were strongly reheated during impact (Nininger 1956, Lipschutz and Anders 1961), and these specimens are also distinguished by an exceptionally low He^3 content (Signer 1961b). Undoubtedly, part of this effect was caused by shielding, but some diffusion loss is also conceivable. This would again lead to an underestimate of the cosmogenic K^{40} content.

In an attempt to explain the high Ar^{40} contents of these meteorites, Marshall (1962) assumed that solid objects formed long before the formation of the solar system, and that the iron meteorites were derived from these objects by some reworking 5 Gyr ago. The largest masses or those heated for only a very short time would be most likely to retain some of the radiogenic Ar^{40} formed prior to the formation of the solar system.

A somewhat more likely possibility is the trapping of primordial or radiogenic Ar^{40} in the meteorite parent body. At least two iron meteorites are known to contain primordial He^4 (*vide infra*), and, if the meteorite parent body had been sufficiently gas-tight to retain an "internal atmosphere" (Sec. 7), the solidification of the iron meteorites would have proceeded under a finite partial pressure of noble gases, including Ar^{40}. For the achondrite Pesyanoe, with an Ar^{36} content of 159×10^{-8} cc (at STP)/g, Gerling and Levskii deduced a partial pressure of argon of 0.9 mm Hg (and a total noble-gas pressure in the gas phase of $\sim$2200 mm Hg). Assuming equal solubilities of noble gases in silicates and metal, and an Ar^{36}/Ar^{40} ratio of 1 in the primordial gas comprising the "internal atmosphere," one finds that an Ar^{40} partial pressure of 5×10^{-3} mm Hg, and a total noble-gas pressure of <0.2 atm would be required to account for the highest Ar^{40} content observed by Stoenner and Zähringer in the meteorite with the lowest content of cosmogenic gases. Even if *no* primordial Ar^{40} had been trapped in the body, it is found that the required partial pressures of 5×10^{-3} mm Ar^{40} (at the freezing point of nickel-iron) would be produced by the decay of K^{40} in 35 Myr, for an assumed volume of the internal atmosphere of 5%. This time is of the same order of magnitude as the cooling times derived from other considerations (Table XIV, Sec. 6).

There is thus considerable doubt concerning the content of radiogenic Ar^{40} and noncosmogenic K^{40}. For several other irons not dated by Stoenner and Zähringer, Voshage and Hintenberger (1961) have measured the isotopic composition of potassium, and found the abundances of K^{39}, K^{40}, and K^{41} to be roughly in the ratio 2:1:2. This ratio is so different from the normal terrestrial one of $10:10^{-3}:1$ that only very minor amounts of normal potassium can be present in these meteorites. Obviously, the presence of normal potassium and of radiogenic argon must be proven conclusively before the feasibility of K-Ar dating of iron meteorites can be regarded as established.

5.2 U-He Ages

The first attempts to measure meteorite ages involved the uranium-helium method (Paneth, Gehlen, and Guenther 1928; Arrol, Jacobi, and Paneth 1942). Unfortunately, all of the early results obtained by this method were in error for two reasons: nearly all the helium in iron meteorites is cosmogenic rather than radiogenic (Bauer 1947, 1948; Paneth, Reasbeck, and Mayne 1952; Singer 1952, 1957; Öpik and Singer 1957) and the true uranium content of iron meteorites, as measured by neutron activation analysis, is far lower ($3\times10^{-3}-0.3$ ppb, Reed and Turkevich 1955; Reed, Hamaguchi, and Turkevich 1958) than the apparent contents measured by wet chemical analysis or other techniques (10–100 ppb; Davis 1950; Dalton, Golden, Martin, Mercer, and Thomson 1953; Dalton, Paneth, Reasbeck, Thomson, and Mayne 1953; Dalton and Thomson 1954; Deutsch, Houtermans, and Picciotto 1956).

The ataxites Washington County and Tucson contain anomalously high amounts of He^4 ($He^3/He^4 = 0.067$ and 0.07, compared to "normal" values of $0.25-0.3$). This overabundance might be attributed either to radioactive decay of U and Th, or to occlusion of primordial noble gases (Schaeffer and Fisher 1959; Fisher and Schaeffer 1960). The former possibility is now ruled out, since the uranium contents of these meteorites are too low to account for the observed amounts of He^4 (Reed 1961).

It thus appears that no iron meteorites contain sufficient amounts of U and Th to permit dating by this method. However, some troilites have uranium contents in the range 1–10 ppb (Reed, Kigoshi, and Turkevich 1960; Goles and Anders 1961c), and the (U,Th)-He method may yet prove applicable to iron meteorites of sufficiently low cosmogenic-helium content. Also, the silicate inclusions other than olivine, which are found in some irons (Kodaikanal, Weekeroo Station), may contain appreciable amounts of uranium.

For chondrites, the situation is far more favorable. The He^3/He^4 ratio is typically the order of 10^{-2}, so that the correction for cosmogenic He^4 is usually less than 10%. The uranium content of chondrites seems to be rather constant at 11 ppb (Hamaguchi, Reed, and Turkevich 1957), and the Th/U ratio varies little from the mean of 3.6 (Bate, Huizenga, and Potratz 1959). Reed and Turkevich (1957) have, therefore, calculated (U,Th)-He^4 ages for a number of chondrites of known helium content, using the above mean values for the U content and the Th/U ratio. This practice was also followed by Eberhardt and Hess (1960). For greater accuracy, however, the actual uranium and thorium contents should be measured, since recent work indicates that the dispersion in the uranium contents of chondrites, particularly among the rarer subclasses may be somewhat greater than originally assumed (Reed, Kigoshi, and Turkevich 1960; Goles and Anders 1961c).

The calcium-rich achondrites usually have higher uranium contents, the order of 10^2 ppb, and a large proportion of their helium appears to be radiogenic. Unfortunately, measurements of both He and U are available for only four meteorities (Table XIII).

The uranium content of calcium-poor achondrites and pallasites is not well known but appears to be at or below the value for chondrites. Judging from the limited data available (Eberhardt and Hess 1960; Hintenberger, König, and Wänke 1961), the correction for cosmogenic He^4 may exceed 50%, so that the uranium method does not hold very much promise for these meteorites.

5.3 Interpretation

The following trends are evident in Tables XII and XIII:

(1) Both the K-Ar and the (U,Th)-He ages show real and large variations, ranging from 0.37 to 4.5 Gyr. This variation stands in sharp contrast to the clustering of the solidification ages near 4.6±0.2 Gyr.

(2) When both K-Ar and (U,Th)-He ages are measured for the same meteorite, the values agree fairly well in the majority of cases (21), though in seven instances large discrepancies occur (Fig. 9). At least four of these cases, (Nuevo Laredo, Beddgelert, Breitscheid, and Pultusk) are well substantiated, and in the case of St. Michel only the uranium content has not been directly measured.

(3) When two components of a polymict breccia are dated separately, the dates do not agree (e.g., Kunashak and Pervomaiskii). From the very limited evidence available, this appears to be true for both the K-Ar and the (U,Th)-He methods.

In the very simplest interpretation, these ages are assumed to give the date of the final cooling of the meteorite, following a high-temperature era during which no retention of radiogenic gases was possible. By analogy with terrestrial rocks, this high-temperature era is assumed to coincide with the formation time of the meteoritic minerals.

Unfortunately, this simple model of rapid, monotonic cooling cannot explain observation (3) and the seven exceptional cases under (2), in the foregoing. A discrete heating event should result in identical K-Ar and (U,Th)-He ages for a given meteorite, and for the components of a polymict breccia. Moreover, it is already extraordinarily difficult to conceive of a satisfactory heat source for melting the meteorites just once in their history. (Urey 1955, 1956b, 1957a, b, 1958; Fish, Goles, and Anders 1960). To demand as many episoidic heating events as there are ages only compounds these difficulties.

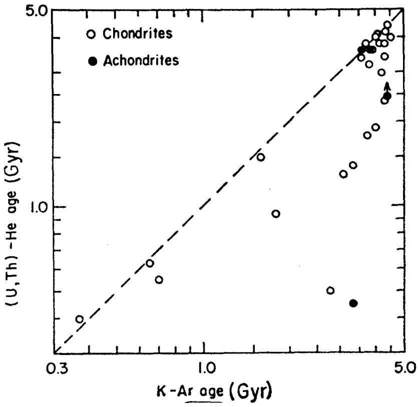

FIG. 9. $K^{40}-Ar^{40}$ vs $(U,Th)-He^4$ ages of stone meteorites. The agreement is good in the majority of cases, though in 7 instances the discrepancy is large enough to suggest preferential loss of He^4. The (U,Th)-He age for Norton County is a lower limit.

However, it is not necessary to heat the meteorites to their melting point in order to expel the radiogenic gases. Large gas losses can occur even at low temperatures and it is, therefore, possible to formulate a more attractive model based on continuous diffusion losses. Stoenner and Zähringer (1958), Geiss and Hess (1958), and Eberhardt and Hess (1960) mentioned long-term diffusion losses as a possible explanation of the short gas-retention ages. This problem has been treated in greater detail by Goles, Fish, and Anders (1960).

Taking the solidification ages (~4.5 Gyr) as the actual formation times of the meteoritic minerals, Goles *et al.* pointed out that there are three consecutive eras during which gas losses can occur[8]: 1. cooling of the meteorite parent bodies; 2. "storage" of the meteorite in the parent bodies at high enough temperatures to allow some diffusion losses to take place; 3. solar heating of the meteorite after breakup of the parent body.

We consider first the limiting case where the entire diffusion loss takes place in the initial cooling of the meteorite parent bodies. It can be shown that Ar^{40} retention by meteorites requires temperatures well below 300°K. Hence, the meteorite parent bodies must have cooled to 300°K or less, before Ar^{40} could begin to accumulate in the meteorites-to-be. These cooling times are an extremely sensitive function of the size of the parent body, as shown in Table XIV (Allan and Jacobs 1956; Goles and Fish 1961). Thus, the relatively

---

[8] The discussion is given in terms of the K−Ar method, but the results are, of course, pertinent to the (U, Th)−He method as well.

TABLE XIV. Cooling times for asteroids initially melted 4.5 Gyr ago.[a] (Goles and Fish 1961).

Radius	Time
km	Gyr
700	5.6
500	4.8
375	1.8
250	0.48
180	0.15
100	0.04

[a] Time required to cool from a mean temperature of 1000°K to 300°K at a radiation temperature of 105°K.

small differences between the majority of the K-Ar ages and solidification ages are most simply explained in terms of sizes of no more than a few hundred kilometers for the meteorite parent bodies. If the meteorites were to have originated in larger bodies, either of two additional assumptions must be made: The chondrites come only from cool surface regions of this body, or the body broke up gradually, each K-Ar age signifying the date of one such breakup. Of course, in either case subsequent breakup to meter-sized fragments must be postulated to account for the short cosmic-ray-exposure ages of the chondrites.

For the next limiting case, we assume that the parent bodies cooled relatively rapidly and that the entire diffusion loss took place during storage of the meteorite-to-be in its parent body. The extent of the losses, of course, depend on the temperature within the body. It should be possible to specify two limiting temperatures T_1 and T_2 such that diffusion losses are virtually zero below T_1, and nearly complete above T_2. Between these two limits, partial losses can occur.

In case of diffusion losses, an apparent age t^* is observed instead of the true age t. In the somewhat idealized model of Goles *et al.*, based on the treatment of Wasserburg (1954) t^* can be expressed as a function of t, the decay constant λ, and the diffusion parameter D/a^2 (D=diffusion coefficient; a=grain radius)

$$t^*=\frac{1}{\lambda}\ln\left[1+\sum_{n=1}^{\infty}\frac{\lambda}{\lambda-Dn^2\pi^2/a^2}\cdot\frac{6}{n^2\pi^2}\times\left\{\exp\left(\lambda t-\frac{Dn^2\pi^2 t}{a^2}\right)-1\right\}\right]. \quad (20)$$

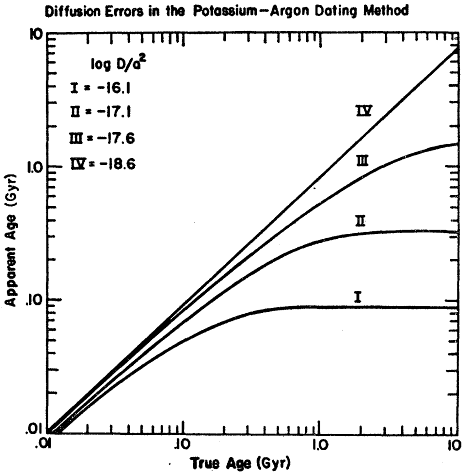

FIG. 10. True vs apparent $K^{40}-Ar^{40}$ age. (Goles, Fish, and Anders 1960).

Figures 10 and 11 show plots of t^* vs t calculated from Eq. (20). For meteorites with a true K-Ar age of 4.5 Gyr, measurable Ar losses occur even if D/a^2 is as small as 1×10^{-20} sec^{-1}, and, at $D/a^2=2.5\times10^{-17}$ sec^{-1}, the rates of argon production and argon loss balance each other at an Ar^{40}/K^{40} ratio so low as to give an apparent age of 0.09 Gyr for any true age greater than 0.4 Gyr.

For a quantitative treatment of these diffusion losses, it is essential to establish the relation between temperature and D/a^2 for meteorites. Extrapolating the limited literature data to low temperatures, Goles *et al.* concluded that K-Ar ages of >4 Gyr implied mean storage temperatures of $\leq190\pm30$°K. Even for objects as "young" as Ramsdorf (0.37 Gyr), storage temperatures of only ~230°K are required. All intermediate ages could, in principle, be attributed to storage at intermediate temperatures. The absolute values of these temperatures are, of course, in doubt, but the uncertainties are not likely to be large enough to affect the conclusions significantly.

The heat source for the attainment of these temperatures can be a combination of solar heat and long-lived radioactivity. The temperature of an object heated by solar radiation alone is given by the equation (Drummeter and Schach 1958)

$$T^4=\frac{C}{R^2}\frac{B}{S\delta}\gamma, \quad (21)$$

where C is the solar constant, R is the distance from the Sun in a.u., S is the total surface area of the object, B is its cross-sectional area effective in intercepting solar radiation, δ is the Stefan-Boltzmann constant, and γ is the ratio of solar absorptivity to low-temperature emissivity for the object. A more detailed treatment of this problem has recently been given by Peebles and Dicke (1962). For matter of chondritic composition located in the asteroidal belt, temperatures between 100° and 200°K may be expected. Thus, solar heating alone is nearly sufficient to raise temperatures to the 190°–230°K range. Given only a slight additional amount of heating by long-lived radioactivities, there are many plausible combinations of orbit, radius, and burial depth that will give temperatures of this order.[9]

---

[9] Even the higher of these values allows for only a slight temperature increase due to heating by long-lived radioactivities. This constitutes a second and wholly independent argument in favor of asteroidal-sized parent bodies less than 250 km in radius. Of course, just as in the previous section, it is again possible to devise more complicated histories involving larger bodies.

The final limiting case assumes all diffusion losses to have taken place *after* the breakup of the parent body, during the cosmic-ray-exposure era of the meteorite. If we regard the meteorites as having originated in the asteroidal belt, then an important distinction exists between the orbit of the parent body and that of the meteorite proper. The parent body is likely to have travelled in a nearly circular orbit with a perihelion between 1.5 and 5 a.u. and most probably near 3 a.u. The meteorite, on the other hand, must have travelled in a highly eccentric orbit with a perihelion of ≤ 1 a.u. It is evident from Eq. (21) that this difference in perihelia corresponds to a substantial increase in temperature. At 1 a.u., the temperature of material of chondritic composition is probably close to the subsurface temperature of the Moon, 234°K (Piddington and Minnett 1949; Goles *et al.* 1960). Although very little diffusion loss takes place until the meteorite approaches perihelion (numerical integration shows that the diffusion loss integrated over the *entire* orbit is approximately that incurred at the perihelion temperature in only 0.025 of the orbital period), the perihelion temperature is high enough even for perihelia lying barely within the Earth's orbit to give noticeable diffusion losses for chondrites with cosmic-ray-exposure ages as short as 10^7–10^8 yr.

An example calculated by Goles *et al.* predicts an apparent K-Ar age of 4.39 Gyr for a true K-Ar age of 4.55 Gyr, an exposure age of 2.4×10^7 yr, and a perihelion distance of 0.95 a.u. Recently, it has become possible to check this calculation for gross inconsistencies in at least one case. For the Příbram chondrite, the only one with a photographically determined orbit, the perihelion was 0.79 a.u. Hence, for an exposure age of 2.2×10^7 yr and a "true" age of 4.5 Gyr, the error in the age should be −0.69 Gyr, corresponding to an apparent K-Ar of 3.9 Gyr. The actual K-Ar age measured by Stauffer (1961b) was 3.6 Gyr, not inconsistent with the predicted value, although this agreement in itself does not prove that the argon loss occurred during cosmic-ray exposure.

The relationship between D/a^2 and T is based on very tenuous evidence at present, and could well be in error by some tens of degrees in the temperature range of interest. Unpublished work by Stauffer (1960) has not revealed any major discrepancies, however, except for some curvature of the log D/a^2 vs $1/T$ plot at low temperatures. This curvature is apparently due to adsorption effects in the laboratory experiment, which slow down the release rate of argon. However, in the near-perfect vacuum of interplanetary space and in the incomparably longer times available for desorption, these effects are not likely to be of any importance. Also, Goles *et al.* have assumed that volume diffusion through the mineral grains is the rate-determining step and that grain-boundary diffusion is likely to be fast even over dimensions of hundreds of kilometers. If this assumption were not valid, the Ar^{40} would remain in the pores of the material instead of escaping from the body. As argued by Goles *et al.*, this "interstitial" Ar^{40} should escape from the meteorite soon after the breakup of the parent body, so that the identical diffusion loss will be observed.

5.3.1 *Origin of the Diffusion Losses*

Thus, a large ambiguity exists: Did the gas loss occur mainly during initial cooling of the parent bodies, during storage, or during the cosmic-ray-exposure era? Obviously, physical conditions during any one of these eras could easily have been such as to account for the entire diffusion loss observed.

Some independent evidence suggests that diffusion losses during the initial cooling stage were appreciable. The I^{129}-Xe^{129} decay intervals (Secs. 6.1 and 6.4) are systematically longer by as much as 10^8 yr than those derived for extinct radionuclides with nonvolatile daughter elements. This implies that temperatures low enough to retain radiogenic Xe^{129} were not reached until some 10^7-10^8 yr after the cessation of melting in the meteorite parent bodies. This temperature is not identical with the temperature at which Ar^{40} is retained, since the diffusion behavior and the mineralogical association of the two gases are different. Further data for many more meteorites will be needed to settle this question, but it seems likely that this factor alone cannot account for some of the short K-Ar ages.

Diffusion losses during storage are plausible inasmuch as objects the size of the larger asteroids are bound to have temperatures of 200°K and higher throughout much of their volume. Even in asteroids of 100-km radius such temperatures can prevail near the center. Diffusion losses at this stage should produce a correlation between the stratigraphic position of a meteorite (as given by its texture and porosity) and its age. Gerling and Rik (1955) claimed to have observed such a correlation, the "recrystallized" or "metamorphosed" chondrites tending to have shorter K-Ar ages than the

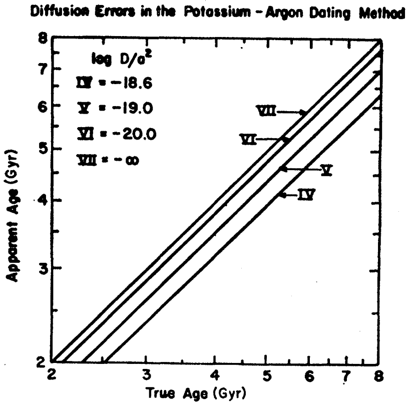

FIG. 11. True vs apparent K^{40}—Ar^{40} age (Goles *et al.* 1960). Even at D/a^2 values as small as 2.5×10^{-19} sec^{-1} errors as large as 1 Gyr can occur in meteorites with true ages of 4.5 Gyr.

unrecrystallized ones. Also, some additional trends of a very tentative nature can be discerned in Tables XII and XIII: the two shergottites have very short ages and the other Ca-rich achondrites also tend to have somewhat shorter ages than the chondrites. Possibly, this implies an origin in deeper and, therefore, warmer regions of the parent body (Goles *et al.* 1960). The same correlation would result from gas losses during cosmic-ray exposure, if meteorites from similar stratigraphic positions were thrown into orbits with similar perihelia. Indeed, the observed correlation between cosmic-ray ages and K-Ar ages (Table III) gives some support to this interpretation.

The agreement of most K-Ar ages with the (U,Th)-He ages is of decisive importance in this connection. Such agreement would be difficult to obtain by storage in the "intermediate"-temperature range (190–230°K), where only a *partial* loss of argon could occur.

Owing to the differences in half-lives of the parent nuclides, most of the Ar^{40} was produced during the early part of the meteorites' history, whereas much of the He^4 was produced relatively recently. Also, He^4 diffuses much more readily than Ar^{40}, so that, even if their parent nuclides resided in the same mineral (making a the same), the D/a^2 values will differ in both absolute value and temperature dependence. These differences are accentuated if a large proportion of the helium nuclei, having been emitted with considerable kinetic energy, come to rest in a mineral other than the U- and Th-bearing mineral (Turkevich 1961). Finally, the analog of Eq. (20) for the (U,Th)-He ages contains three different λ's so that a rather different dependence of t^* on D/a^2 results. It would be an extraordinary coincidence if all these factors combined to give concordant apparent ages in a single instance; to obtain them in a large number of cases covering more than a tenfold variation in t^* values borders on the impossible.

It seems much more probable that concordant K-Ar and U-He ages significantly shorter than 4.5 Gyr record some discrete event accompanied by an abrupt temperature drop. Prior to this event, the temperature must have been high enough to result in virtually complete loss of both He^4 and Ar^{40}, and, after this event, the temperature must have fallen sufficiently fast to permit complete retention of both gases.

An event most closely meeting these criteria is a collisional breakup. To account for the observations, this breakup must have shattered an object large enough to have a warm interior due to radioactive heating, i.e., ≥ 100 km in radius, to fragments small enough to be opaque to cosmic rays (100 km$>r\geq$10m).

The "short" Ar^{40} and He^4 ages only give a lower limit to the temperatures prevailing in this object prior to the breakup: they must have been high enough to cause complete loss of these gases. But, it is also possible to infer an upper limit to this temperature. The Bruderheim meteorite, with a K-Ar age of 1.6 Gyr, has, nevertheless, retained some radiogenic Xe^{129} from the decay of extinct I^{129} (Krummenacher, Merrihue, Pepin, and Reynolds 1961; Tables XV and XVI of this paper). This Xe^{129} was generated in the meteorite as early as ~4.5 Gyr ago. Although it is known that Xe^{129} diffuses from meteorites somewhat less readily than Ar^{40} (Zähringer and Gentner 1961; Jeffery and Reynolds 1961a), the temperature at which Xe^{129} would be lost completely can hardly be higher than 300–400°K. Thus, the thermal history of Bruderheim suggested by these data involves an initial cooling from melting temperatures, ~4.5 Gyr ago, to <300–400°K within $\leq 10^8$ yr, storage at temperatures between ~240° and 300–400°K for the next 3 Gyr, and finally, an abrupt temperature drop to $\leq$190°K some 1.6 Gyr ago. This history could be made much more definite if more diffusion data for Ar^{40} and Xe^{129} were available.

The discordant argon and helium ages (e.g., Nuevo Laredo, St. Michel, Beddgelert, etc.) require a different history: either storage at "intermediate" temperatures near 200°K where argon losses are slight, while helium losses are large, or reheating during the cosmic-ray-exposure era. No definite choice between these alternatives appears possible at the moment.

6. EXTINCT RADIOACTIVITY

In 1947, Harrison Brown pointed out the possibility that "extinct" radionuclides with half-lives of 10^8 yr or less might once have been present in the early solar system, if nucleogenesis had taken place shortly before its formation (Brown 1947). Such radioactivities could be of twofold significance for the meteorites. They might provide the basis for new dating methods which, because of the short half-lives involved, would be of high relative accuracy. But, they might also have had important effects on the thermal history of the meteorite parent bodies (Urey 1955; Fish, Goles, and Anders 1960) and of the Earth (Rosenblatt 1953; Kohman 1953, 1956). The early literature on the subject is well reviewed in Kohman's 1956 paper.

The equations used in this dating method are derived as follows. We suppose that at some time in the past the extinct radionuclide X^A was synthesized in the galaxy and became incorporated in the solar nebula. This nuclide decays with a mean life τ to the stable nuclide Y^A. At some time t_0, production of X^A ceased and unsupported decay began. Let the initial amounts at t_0 be designated $X_0{}^A$ and $Y_0{}^A$. After the formation of solid objects, chemical fractionations between elements X and Y set in. If the last such fractionation occurred at time t and the object remained a closed system thereafter, the one-time abundance of X^A can be related to the present-day abundance of Y^A. Defining $\Delta t_{XY}\equiv t-t_0$, we write

$$X_t{}^A = X_0{}^A e^{-(\Delta t_{XY}/\tau)} = Y_{r,p}{}^A = Y_p{}^A - Y_0{}^A - Y_{r,t}{}^A, \quad (21)$$

where r=radiogenic and p=present. For convenience, the abundances can be normalized to the stable, nonradiogenic isotopes X^B and Y^C

$$\frac{X_t^A}{Y^C}=\frac{X_0^A}{X^B}\times\frac{X^B}{Y^C}e^{-(\Delta t_{XY}/\tau)}=\frac{Y_{r,p}^A}{Y^C}=\frac{Y_p^A-Y_t^A}{Y^C}, \quad (22)$$

where

$$Y_t^A=Y_0^A+Y_{r,t}^A.$$

This involves the tacit assumption that the solar nebula was isotopically homogeneous and that no appreciable isotopic fractionations took place (other than those caused by radioactive decay).

In principle, this equation can be used to calculate Δt_{XY}, but there are two practical difficulties in the application of this method. The amount of X^A at t_0 (i.e., X_0^A/X^B) and the isotopic composition of Y at time t (specifically, Y_t^A/Y^C) must be known. The latter can hardly even be determined exactly, since it requires, for every object to be dated, another object that separated from the solar nebula at the identical time t, with so high a ratio of Y to X that subsequent decay of X^A did not change the isotopic composition of Y appreciably. Under favorable circumstances (e.g., when X_0^A/Y_0^A is small and X_t^A/Y_t^A is large), even a crude approximation of Y_t^A/Y^C will suffice, but when the isotopic anomaly is small the uncertainties become quite large. To stress the distinction between Y_0^A/Y^C, which may be regarded constant for the entire solar system, and Y_t^A/Y^C, which varies with t, we use the terms "primordial" and "primeval" for these two quantities.

The amount of the extinct radionuclide present at t_0 depends on the model assumed for nucleosynthesis. The simplest model, which also happens to be the only one considered in the earlier literature on the subject, assumes that all nuclides now present in the solar system were synthesized in a single event ("sudden synthesis"). Such an event should produce extinct and stable nuclides in comparable proportions, and the initial abundances of extinct radionuclides can, therefore, be estimated by simple interpolation of the semiempirical "solar-system" abundance curve (Suess and Urey 1956; Cameron 1959), or from more elaborate types of nuclear systematics (Burbidge, Burbidge, Fowler, and Hoyle 1957). The abundance ratio of two nuclides X^A/X^B should simply equal the ratio of the two production rates K^A/K^B. Substituting this ratio in Eq. (22), we obtain

$$\frac{Y_p^A-Y_t^A}{Y^C}=\frac{K_A}{K^B}\cdot\frac{X^B}{Y^C}\cdot e^{-\Delta t_{XY}/\tau}. \quad (23)$$

The one-time presence of extinct radionuclides with half-lives of $\sim 10^7$ yr in meteorites implies, by the "sudden-synthesis" model, that the synthesis of the extinct nuclides and all other matter occurred shortly, perhaps $\leq 10^7-10^8$ years, before the formation of the meteorites. This notion is difficult to sustain in the face of evidence for continuous production of heavy nuclei in stars (Burbidge *et al.* 1957). Hence, a second model involving continuous nucleosynthesis throughout the Galaxy must be considered. In barest outline, the processes envisaged in this model are the following. Heavy nuclei are continuously synthesized in supernovae and red giants by a variety of processes (*r* process, neutron capture on a fast time scale; *s* process, neutron capture on a slow time scale; *e* process, high-temperature synthesis under conditions of thermodynamic equilibrium; etc.). The products are returned to the interstellar medium through a variety of ejection mechanisms, and are recycled by incorporation in newly forming stars. Assuming perfect mixing, the interstellar medium should thus always contain extinct radionuclides in steady-state proportions. To estimate these proportions, one must make additional assumptions about the over-all rate of nucleosynthesis in the Galaxy.

The simplest variant of this model assumes a constant rate throughout the lifetime of the Galaxy (Wasserburg, Fowler, and Hoyle 1960). In that case, the amount of a stable nuclide X^B synthesized during time T (defined as the duration of nucleosynthesis prior to the formation of the solar system) equals

$$X^B=K_BT.$$

For a radioactive nuclide X^A, the corresponding equation is

$$X^A=K_A\tau(1-e^{-T/\tau}).$$

In the special case of an extinct radionuclide with $\tau\ll T$, this equation simplifies to

$$X^A\approx K_A\tau \quad (\text{for } \tau\ll T).$$

Hence, the ratio X_0^A/X^B in Eq. (22) equals

$$\frac{X_0^A}{X^B}=\frac{K_A}{K_B}\cdot\frac{\tau}{T} \quad (24)$$

and

$$\frac{Y_p^A-Y_t^A}{Y^C}=\frac{K_A}{K_B}\cdot\frac{X^B}{Y^C}\cdot\frac{\tau}{T}\cdot e^{-\Delta t_{XY}/\tau}. \quad (25)$$

Solving for Δt_{XY}, we obtain

$$\Delta t_{XY}=\tau\ln\left\{\frac{Y^C}{Y_p^A-Y_t^A}\cdot\frac{K^A}{K^B}\cdot\frac{X^B}{Y^C}\cdot\frac{\tau}{T}\right\}. \quad (26)$$

This model may be an oversimplification inasmuch as the rate of nucleosynthesis can hardly have been constant over the lifetime of the Galaxy. Not only will stellar evolution rates change with time, owing to the gradually increasing metal content of newly formed stars but massive stars evolve very rapidly also, and the proportion of such stars must have decreased with time. Hence, the rate of nucleosynthesis should have

TABLE XV. Xenon in meteorites and the earth abundance relative to Xe^{130}.

Object	Class	Xe content g Xe/g	124	126	128	129	131	132	134	136	Reference
Mighei	K		0.0299	0.0271	0.521	6.71+0.05	5.11	6.32	2.38	2.01	b
Murray	K	2.5×10^{-10}	0.0295	0.0270	0.510	6.48±0.04	5.04	6.14	2.34	1.97	b
Orgueil	K		0.0311	0.0270	0.518	6.62±0.05	5.10	6.23	2.37	2.01	b
Abee	Cek		···	···	···	15.2 ±0.6	5.02	6.35	2.37	1.98	c
Indarch3	Cek	4×10^{-11}	0.0265	0.0219	···	19.1 ±1.1	4.87	6.05	2.35	1.98	b
Indarch 4	Cek		0.0268	0.0239	···	21.6 ±0.1	5.08	6.28	2.45	2.10	b
St. Marks	Cek		0.0276	0.0229	···	20.0 ±0.4	5.02	6.11	2.38	2.00	b
Bruderheim	C		0.0293	0.0310	···	7.49±0.07	5.13	6.39	2.50	2.15	b
Bruderheim		6.1×10^{-12}	0.0310	0.0321	···	7.37±0.05	5.01	6.23	2.45	2.18	d
Elenovka	Ck (?)		···	···	0.488	6.96±0.23	5.22	6.47	2.50	2.13	b
Kyushu	Cwa		···	···	···	6.65±0.09	5.07	6.35	2.45	2.06	b
Richardton	Cca	6.1×10^{-12}	0.0339	0.0287	0.525	8.99±0.05	5.06	6.17	2.39	1.99	b
Sardis (metal)	Ogg	1.5×10^{-13}			···	7.53±0.16	5.36				d
Sardis (troilite)	Ogg	2.5×10^{-12}			0.68	8.54±0.06	5.62				d
Earth[a]	Cca	3.8×10^{-13}	0.0236	0.0220	0.471	6.49±0.03	5.20	6.60	2.56	2.18	f

[a] Atmospheric xenon only.
[b] Krummenacher, Merrihue, Pepin, and Reynolds (1961).
[c] Reynolds (1960c).
[d] Clarke and Thode (1961).
[e] Reynolds, Merrihue, and Pepin (1962).
[f] Goles and Anders (1961a).

declined with time as more and more matter became locked up in small and slowly evolving stars. To allow for this effect, Kohman (1961) proposed inclusion of another negative exponential term in Eq. (25), whereas Cameron (1961) simply uses a longer effective T (20 instead of 10 Gyr).

The third model, "mixed nucleosynthesis," attempts to combine the two preceding ones by assuming that, in addition to galactic synthesis, some local production in the solar neighborhood took place shortly before the formation of the meteorite parent bodies. This local event might have been a nearby supernova (Kohman 1961; Murthy and Urey 1962) or charged-particle acceleration during the formation of the solar system (Fowler 1960; Cameron 1960).

Which of these models best represents the true situation can only be decided after a careful study of the evidence. As a point of departure, the "continuous-synthesis" model of Wasserburg *et al.* is particularly well suited. It provides a self-consistent set of Δt's for each extinct radionuclide. From the degree of agreement of these sets of Δt's one can infer the shortcomings of the model and the modifications required to bring the Δt's into accord.

The time interval measured by this method is not an "age" in the sense of the K-Ar or Pb-Pb ages, since it is measured not backward in time from the present but forward in time from some elusive, but nevertheless definable, starting point t_0. To emphasize the distinction between these dates and "ages" properly so called, two new terms have been proposed: "formation intervals" (Cameron 1960, Reynolds 1960c) and "decay intervals" (Goles and Anders 1960). The former term rightly stresses the fact that the formation of the meteorite parent bodies must have taken place during this interval, but is misleading inasmuch as this formation process is not directly linked to the "formation interval": neither its beginning nor its end coincide with the corresponding points of the formation interval. The term "decay interval" stresses the point that unsupported decay of I^{129}, without chemical fractionation and without retention of the decay product, is the actual physical event measured by this method.

6.1 Iodine-129

The β^- emitter iodine-129 is the extinct radionuclide *par excellence.* Its half-life, 16.4 Myr (Kohman 1961), lies in a convenient range, its abundance is easy to estimate, and its decay product, Xe^{129}, should be easy to detect even at low levels since the meteorites (and the Earth) seem to have retained very little primordial xenon.

After two earlier, unsuccessful searches (Wasserburg and Hayden 1955b; Reynolds and Lipson 1957), radiogenic Xe^{129} from the decay of I^{129} was at least detected by Reynolds (1960a) in the Richardton chondrite. This discovery was soon confirmed (Signer 1960) and extended to other meteorites (Reynolds 1960b, c, d; Zähringer and Gentner 1960; Krummenacher, Merrihue, Pepin, and Reynolds 1961). The data are shown in Table XV.

On the basis of their xenon content, the meteorites fall into three classes. The "ordinary" (bronzite and

TABLE XVI. $I^{129}-Xe^{129}$ decay intervals.

Object	Xe^{129}/Xe^{130}	Xenon content cc/g (at STP)$\times 10^{-11}$ Total	Xenon content cc/g (at STP)$\times 10^{-11}$ Xe_r^{129}	I^{127} ppb	Δt million years	Reference
Beardsley	$\sim 6.5 \pm 2.0$	≤ 4	≤ 1.3	62 ± 11	≥ 190	a, b
Bruderheim	7.37 ± 0.05	104	3.6 ± 0.2	16 ± 6	123 ± 8	b, c
Indarch	19.1 ± 1.1	650	250 ± 100	270 ± 45	91^{+12}_{-8}	b, d
Murray	6.48 ± 0.04	4300	0 ± 11	230 ± 50	≥ 144	b, e
Richardton	8.99 ± 0.05	104	10 ± 1	28 ± 4	113 ± 4	b, f
Sardis troilite	8.54 ± 0.06	42	3.3 ± 0.1	3590 ± 380	254 ± 3	b, g
Earth's atmosphere	6.49 ± 0.03	65	(<17)	(65)	≤ 170	h

[a] Wasserburg and Hayden (1955b).
[b] Goles and Anders (1960, 1961c).
[c] Clarke and Thode (1961).
[d] Reynolds (1960c).
[e] Reynolds (1960b).
[f] Reynolds (1960a).
[g] Reynolds, Merrihue, and Pepin (1962).
[h] Goles and Anders (1961a).

hypersthene) chondrites have low xenon contents and zero to moderate Xe^{129} enrichment. The enstatite chondrites have intermediate xenon contents and very large Xe^{129} anomalies. Finally, the carbonaceous chondrites have very large xenon contents with only a slight, if any, Xe^{129} enrichment.

Particularly for meteorites with slight Xe^{129} anomalies, a large ambiguity arises from the correction for primeval xenon. For quantitative interpretation, the mass spectrum is usually normalized to some reference isotope Xe^j, and expressed in the form of isotopic ratios Xe^i/Xe^j. The anomalies Δ_i can then be found by subtracting the corresponding ratios of the primeval-xenon spectrum. In practice, atmospheric xenon is often used as an approximation of primeval xenon:

$$\Delta_i = \left[\frac{Xe^i}{Xe^j}\right]_M - \left[\frac{Xe^i}{Xe^j}\right]_T,$$

where the subscripts M and T stand for "meteorite" and "terrestrial."

Some question might be raised about the appropriateness of atmospheric xenon as an approximation of primeval xenon. Various types of isotope systematics have been used to estimate the amount of a radiogenic component in atmospheric Xe^{129} and values ranging from $\leq 20\%$ (Katcoff, Schaeffer, and Hastings 1951) to $\sim 87\%$ (Cherdyntsev 1961) have been proposed. Decay of I^{129} after the onset of I-Xe fractionations in the solar nebula might, of course, have given rise to local variations in the Xe^{129} isotopic abundance of primeval xenon, and the Xe^{129} abundance in atmospheric xenon could have been further altered by decay of I^{129} in the Earth itself. It seems, however, that such effects were small: Murray xenon and terrestrial xenon have virtually the same Xe^{129}/Xe^{130} ratio (Table XV) in spite of the great differences in the location and in the xenon and iodine contents of the two objects.

Unfortunately, as shown by Reynolds, the mass spectra of meteoritic and terrestrial xenon are incongruent at all mass numbers, not just at mass 129. Hence, the apparent anomalies will change in magnitude and even sign, depending on the choice of reference isotope for normalization. Reynolds normalized his data to Xe^{136}, but, as Kuroda (1960) pointed out, the abundance of this nuclide could have been altered during geologic time, since it is a prominent fission product of uranium and extinct transuranium elements. Kuroda, therefore, chose to normalize the data to the shielded isotope Xe^{130}, a practice which is also being followed in this paper. Finally, Cameron (1961) proposed normalization to another shielded isotope, Xe^{128}. Though only half as abundant as Xe^{130} (1.92% vs 4.1%), it has the advantage of not being producible from an abundant xenon isotope by neutron capture, a process that Cameron believes to have affected solar and terrestrial xenon. This advantage is largely cancelled by the fact that Xe^{128} can be produced from stable I^{127} by the reactions

$$I^{127}(n,\gamma)I^{128} \xrightarrow[25m]{\beta^-} Xe^{128}.$$

Assuming that the Xe^{129} anomaly arose from the *in situ* decay of I^{129}, one can calculate I^{129}-Xe^{129} decay intervals for those meteorites whose xenon and iodine contents are known (Table XVI). The following parameters are used in this calculation:

1. $T = 1 \times 10^{10}$ yr.
2. $Xe^{129}/Xe^{130} = 6.48$ (mean of the ratio in Murray and Earth's atmosphere). The appropriateness of this ratio is open to question (see Sec. 6), but the error introduced is likely to be small at least for meteorites with large anomalies.
3. $K_{129}/K_{127} = 1.0$.
4. $\tau_{129} = 23.6$ Myr.

Only for four of these meteorites (Bruderheim, Indarch, Richardton, and Sardis) are the dates reasonably free from ambiguity. Their Xe^{129} and iodine contents are well determined, and the Xe^{129} anomalies are so large that the correction for primeval xenon need not be known with great accuracy. It is somewhat intriguing that the decay intervals for the two ordinary chondrites Richardton and Bruderheim and for the enstatite chondrite Indarch do not differ greatly from one another, in spite of the differences in texture, mineralogy, and trace-element content between these two classes.[10] This would lend support to the view that the meteorites are congenetic, as has always been assumed in the derivation of the Pb^{207}-Pb^{206} dating method. Patterson (1956) has proved this assumption to be valid within the resolving power of the Pb-Pb method, but the I-Xe method should be able to verify this with greater accuracy.

Of course it must be kept in mind that the two methods do not date the same physical event. The Pb-Pb method dates a U-Pb fractionation, presumably terminating at the time of solidification of the meteorite parent bodies. The I-Xe method dates the onset of Xe^{129} retention by the meteorite-to-be, which could occur only after the temperature fell to some 300°K or below. This cooling time depends not only on the size of the meteorite parent body (Table XIV), but also on the depth of burial within the body. The combination of these two factors could produce a wide spread of I-Xe decay intervals.

It is extremely significant in this connection that the Sardis iron meteorite has a much larger decay interval than the three chondrites just mentioned. It seems that Sardis began to retain radiogenic Xe^{129} some 130–160 Myr later than the chondrites. This difference in relative ages is exactly the opposite of that expected from Urey's theory (1957a, 1958), according to which the chondrites consist of the debris of achondrites and irons, evolved in earlier, "primary" objects of lunar size. On the other hand, it is consistent with the view that the irons come from the cores of internally heated asteroids, which, because of their central location, would cool more slowly than the chondrites in the mantle (Fish *et al.* 1960). Moreover, the cooling times implied by these decay intervals once again indicate a radius near 200 km for the meteorite parent bodies (Table XIV), as already suggested in Sec. 5.3 on the basis of two independent arguments. Of course, it is again possible to devise alternative, *ad hoc* histories involving larger bodies, but the complexity of these hypotheses grows with the amount of evidence to be explained. It is obviously essential to obtain further evidence on the relative decay intervals of irons and stones.

For the remaining two meteorites, large ambiguities exist. The Xe^{129}/Xe^{136} ratio in Murray is 3.28, compared to 2.98 for the atmosphere. Taking atmospheric xenon to represent primeval xenon congenetic with Murray, one can thus compute a radiogenic Xe^{129} content of $1.09\pm0.11\times10^{-9}$ cc (at STP) and a decay interval of $106(+9, -6)$ Myr. However, if Kuroda's normalization is used, the Xe^{129}/Xe^{130} ratios are 6.48 and 6.49, corresponding to a radiogenic Xe^{129} content of $0.00\pm0.11\times10^{-9}$ cc (at STP), and a lower limit of ≥144 Myr for the decay interval.

In (Sec. 6.2) it is shown that the Xe^{130} normalization is preferable. Then, the lack of radiogenic Xe^{129} in Murray and its low K-Ar age (1.9 or ≤2.77 Gyr, Table XII) would imply a long cooling time. However, a prolonged high-temperature era is difficult to reconcile with the presence of primordial noble gases in this meteorite (Reynolds 1960b); Stauffer 1961a). A more satisfactory assumption would be that the radiogenic Ar^{40} and Xe^{129} are located in sites of lower retentivity than the primordial Ar^{36}, Xe^{124}, etc. Then, a very mild heating would cause the loss of the former while permitting retention of the latter. In the case of a few other meteorites at least, such differences in retentivity have indeed been demonstrated for Ar^{36} and Ar^{40} (Geiss and Hess 1958; Zähringer and Gentner 1961) as well as for Xe^{129} and Xe^{132} (Jeffery and Reynolds 1961a). Whichever explanation is finally accepted for Murray, probably applies to the other carbonaceous chondrites as well. Their xenon contents and isotopic compositions are very similar (Reynolds 1960c, Krummenacher *et al.* 1961), as are their iodine contents (Goles and Anders 1961c) and K-Ar ages (Stauffer 1961a).

For Beardsley, two other explanations have been suggested (Goles and Anders 1961a). This meteorite is much more compact than Richardton and may well have originated in a deeper region of its parent body where the effective cooling time would be longer. On the other hand, the Beardsley sample measured by Wasserburg and Hayden (1955b) had been exposed to ground water for at least one year, and, since iodine in "ordinary" chondrites is known to reside in a water-soluble phase (Goles and Anders 1961b), the iodine (and any *in situ* produced Xe^{129}) may well have been lost during terrestrial weathering along with other water-soluble trace elements (Sec. 4.1). A remeasurement on an unweathered Beardsley sample would settle this question.

For the other ordinary chondrites in Table XV, I-Xe decay intervals cannot be calculated, since their absolute xenon contents are not known. In some meteorites (Elenovka and Kyushu), the Xe^{129} anomaly seems to be slight or nonexistent, but since both the xenon and the iodine contents of chondrites can vary considerably (Goles and Anders 1961c) and since the measured isotopic composition is surely affected by varying levels of atmospheric contamination, not even a qualitative comparison seems possible at present. To a lesser extent, this is also true for the remaining enstatite chondrites. Even in the case of these meteorites, with their relatively high xenon contents, atmospheric contamination is quite severe. The Xe^{129}/Xe^{132} ratio in Abee was originally reported by Reynolds (1960d) as 2.39 ± 0.09, but in later work, using a different technique, ratios of 5.5 and 5.3 were obtained (Zähringer and Gentner 1961;

---

[10] For Bruderheim, the short K−Ar age indicates Ar^{40} losses (and, hence, presumably some Xe^{129} losses) during storage in the parent body. A correction for this effect would bring the decay intervals even closer together.

Jeffery and Reynolds 1961a). Until this factor is brought under control, quantitative interpretation of the remaining xenon data in Table XV does not seem possible.

Throughout this discussion, it has been tacitly assumed that the Xe^{129} arose from the decay of extinct I^{129} *in the meteorite.* Most other mechanisms, such as production by cosmic rays, were ruled out by Reynolds (1960a). However, Eberhardt and Geiss (1960b) raised an alternative possibility: the decay of I^{129} might have taken place in the solar nebula, long before the formation of any solid bodies. To account for the differences between meteoritic and terrestrial xenon, this model requires that a partial iodine-xenon fractionation took place in the nebula and that the chemical fractionation factor varied in the proto-Earth and proto-asteroidal regions. In other words, mixing must have been incomplete in the solar nebula.

There are two crucial differences between these hypotheses that lend themselves to experimental tests. According to Eberhardt and Geiss, Xe^{129} must have been well mixed with the remaining xenon isotopes before the xenon was incorporated in the meteorite. All xenon in a given meteorite should, therefore, have the same isotopic composition, regardless of the iodine content of the host phase. In contrast, if the Xe^{129} had been produced by decay *in situ*, one would expect to find the Xe^{129} anomaly to vary from phase to phase, in accordance with the iodine content, the primeval xenon content, and the xenon retentivity of the phase.

Evidence favoring the Eberhardt and Geiss hypothesis was reported by Zähringer and Gentner (1961). They heated a fine powder of the Abee enstatite chondrite to progressively increasing temperatures, and found that the Xe^{129}/Xe^{132} ratio in the expelled gas remained sensibly constant at 5.5 over the temperature range 700° to 1350°C. They concluded that both xenon isotopes resided in the same mineral phase.

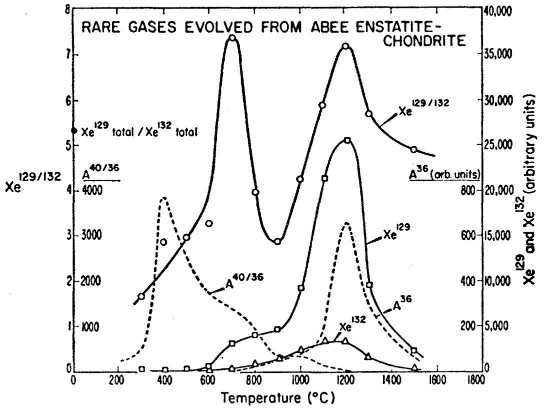

FIG. 12. Evolution of noble gases from Abee meteorite (Jeffery and Reynolds 1961a). Sample heated for 1 hr intervals at temperatures increasing progressively in 100° steps. Note differences in Ar^{40}/Ar^{36} and Xe^{129}/Xe^{132} ratios. These imply different mineralogical associations for radiogenic and cosmogenic argon and for radiogenic and primordial xenon.

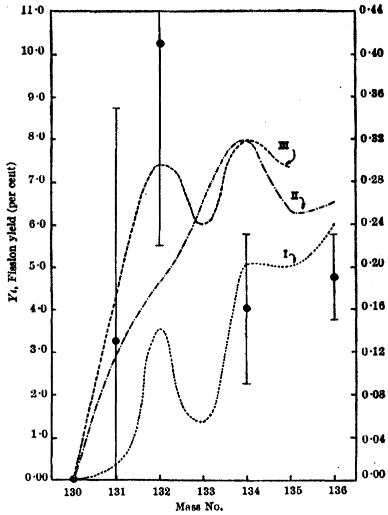

FIG. 13. Xenon anomalies in Earth's atmosphere compared with fission yield curves (Kuroda 1960). Curve I = spontaneous fission of U^{238}, II = neutron-induced fission of U^{235}, III = spontaneous fission of Cm^{242}.

However, Jeffery and Reynolds (1961a) repeated the experiment on the same meteorite and found that the Xe^{129}/Xe^{132} ratio was *not* constant but varied strongly with temperature (Fig. 12). Although it appears that phases rich in Xe^{129} also contain much of the Xe^{132} in the meteorite, this experiment shows that some phases are selectively enriched in Xe^{129} over Xe^{132}, as would be expected for *in situ* decay of I^{129}.

A second, even more conclusive test would be provided by a direct correlation between iodine and xenon-129. If the Xe^{129} had been produced by decay *in situ*, then both I^{127} and Xe^{129} should reside in the same phase, whereas no such correlation, or only an accidental one, would be expected for the hypothesis of Eberhardt and Geiss.

Jeffery and Reynolds (1961b) irradiated a sample of the Abee enstatite chondrite with slow neutrons, thereby converting I^{127} to Xe^{128} *in situ.* The amount and distribution of preterrestrial Xe^{129} was not changed appreciably by the irradiation. Then, by heating the meteorite to progressively higher temperatures, they found that the release of Xe^{129} and Xe^{128} occurred at virtually equal rates over the entire temperature range 500°–1500°C. This proves conclusively that the Xe^{129} is located in the iodine-bearing mineral in this meteorite, and should eliminate the last objections to the soundness of the I^{129}-Xe^{129}-dating method. The nature of the iodine-bearing mineral has not yet been established with certainty. Goles and Anders (1961b) were able to show that iodine is chalcophile and that much of the iodine in chondrites is water-soluble. Both observations point to CaS or a similar sulfide as the likely host mineral. DuFresne and Anders (1961a) have argued that the primordial noble gases in the Pesyanoe meteorite are situated at lattice imperfections in MgS.

TABLE XVII. Thallium in meteorites.[a]

Meteorite	Class	Pb^{204} ppn	Tl ppb	$\frac{Pb^{204}}{Tl^{203}}$	$\frac{Tl^{205}}{Tl^{203}}$	Decay interval (Myr)
Canyon Diablo troilite	Og	98[b]	10.4[b]	32	2.362 ±0.02	≥ 100
Toluca troilite	Om	108[b]	198[b]	1.8	2.365 ±0.02	
Mighei	K	23[b]	140	0.57	2.370 ±0.02	
Abee	Cek	56[b]	150	1.3	2.376 ±0.02	
Beardsley	Cg	2.3[b]	5.8	1.3	2.376 ±0.02	
Richardton	Cca	0.45[c]	0.94	1.6	2.39 ±0.04	
Normal Tl					2.367 ±0.02	

[a] Anders and Stevens (1960).
[b] Reed, Kigoshi, and Turkevich (1960).
[c] Hess and Marshall (1960).

The observed, partial correlation between Xe^{129} and (primordial) Xe^{132} is not in disagreement with these suggestions. (See also Sec. 7)

6.2 Plutonium-244

After the mass spectrum of meteoritic xenon became known, Kuroda (1960) pointed out that the differences between meteoritic and terrestrial xenon seemed to indicate an excess of the heavy isotopes Xe^{131}-Xe^{136} in the Earth's atmosphere. These isotopes happen to be the only ones which would be produced in substantial yield by fission of a heavy nuclide (Khlopin and Gerling 1948). Indeed, when the mass spectra were normalized to Xe^{130}, the Δ_i's showed a striking qualitative resemblance to a typical spontaneous-fission-yield curve (U^{238} or Cm^{242}), although the anomaly at mass 132 corresponded to the improbably high fission yield of $\sim 12\%$ (Fig. 13).

Kuroda showed that the spontaneous fission of U^{238} during the past 4.5 Gyr could account for less than 2% of the observed effect, and suggested that spontaneous fission of extinct Pu^{244} (76 Myr) and Cm^{247} (≥ 40 Myr) or neutron-induced fission of U^{235} (0.71 Myr) might have produced the excess Xe^{131-6}. Indeed, the levels of Pu^{244} expected on the basis of the Wasserburg-Fowler-Hoyle model are more than sufficient to account for all the excess Xe^{131-6} now in the Earth's atmosphere.

Goles and Anders (1961a), therefore, argued that either the Earth must be younger than the meteorites (Pu-Xe decay interval of the Earth = 290 Myr as opposed to I-Xe intervals of 100–120 Myr for the meteorites) or that the solar system received a smaller initial endowment of Pu^{244} than expected from the model of Wasserburg *et al.* (1960). The former view receives some support from the previously mentioned fact that the Earth's atmosphere has virtually the same Xe^{129}/Xe^{130} ratio as the Murray meteorite, although Murray has a thousand-fold-higher xenon content per gram than the Earth. The identity in Xe^{129}/Xe^{130} ratios could, of course, be explained in terms of equal ages and fortuitously identical I/Xe ratios for the two objects, but the explanation in terms of a later origin of the Earth's atmosphere seems less contrived. Still, Cameron (1961) and Kuroda (1961) have shown that the apparent difference in age can be somewhat reduced by a different choice of parameters and a generous allowance for possible errors in these parameters.

Thus far, no evidence has been obtained for fission-produced Xe^{131-6} in meteorites. The levels to be expected are quite low [10^{-11}–10^{-12} cc/g (at STP)], and the detection may be feasible only in meteorites with low primeval xenon content. However, a search for these minute amounts may well be worth the effort, since the fortuitous circumstance that two extinct radionuclides decay to isotopes of the same element, xenon, makes possible the development of a new dating method based on both I^{129} and Pu^{244} (Goles and Anders 1961a). Assuming that $\Delta t_{\mathrm{I\ Xe}} = \Delta t_{\mathrm{Pu\ Xe}}$, i.e., that the iodine- and plutonium-bearing minerals became retentive for xenon at about the same time, one can arrange the decay equations for the two radionuclides in such a way as to eliminate or to determine various unknown quantities, including the primordial I^{129}/I^{127} and Pu^{244}/U^{238} ratios, etc. Thus far, no attempt has been made to apply this method.

6.3 Lead-205

An unsuccessful search for radiogenic Tl^{205} from the decay of ~ 24 Myr Pb^{205} was made by Anders and Stevens (1960). Six meteorites with widely varying Pb/Tl ratios were examined with negative results (Table XVII).

Only in one case was the Pb/Tl ratio high enough to justify calculation of a decay interval. The significance of this value is discussed in Sec. 6.4. But it must be pointed out that the uncertainty in this upper limit is rather large. The production ratio K_{205}/K_{204} is harder to estimate than most ratios of this type, and the half-life of Pb^{205} is not well known either. This nuclide decays mainly, but not exclusively, by L-electron capture, and the present value for the half-life is based on the observed L-capture rate and an *assumed* 20% contribution of electron capture from higher levels. Finally, the Pb^{204} contents of the meteorites analyzed were not measured directly but were taken from the work of Reed, Kigoshi, and Turkevich (1960) and Hess and Marshall (1960). It is possible that the lead contents of some of these meteorites vary significantly from sample to sample.

6.4 Palladium-107

An early search for radiogenic Ag^{107} from the decay of Pd^{107} was unsuccessful (Hess, Marshall, and Urey 1957). They examined silver from a sample of Toluca troilite and found it to be identical with terrestrial silver within ±7%. However, the chemical procedure used in this work is likely to have led to incomplete recovery of silver, thereby increasing the danger of contamination.

When another sample of Toluca troilite was examined by Murthy (1960), he found a 2% enrichment in Ag^{107}, relative to terrestrial silver. Comparable enrichments

TABLE XVIII. Silver in meteorites. (Murthy 1961b).

Sample	Class	$\frac{Ag^{107}}{Ag^{109}}$
Toluca troilite	Om	1.087±0.004
Sikhote Alin	H–Ogg	1.091±0.005
Canyon Diablo	Ogg	1.107±0.003
Reagent $AgNO_3$		1.062–1.066

were later found in several other meteorites (Table XVIII).

Most probably, this enrichment is due to the decay of extinct Pd^{107}. It is not yet possible to calculate accurate decay intervals from these data, since neither the Pd and Ag contents nor the isotopic composition of primeval silver are known. However, it is possible to analyze the data in a way that is less sensitive to errors in the parameters used. From Eq. (25), we find that the abundance of an extinct radionuclide X^A relative to its stable isotope X^B equals

$$\frac{X_t^A}{X^B}=\frac{Y_p^A-Y_t^A}{X^B}=\frac{K_A}{K_B}\cdot\frac{\tau}{T}\cdot e^{-\Delta t_{XY}/\tau}.$$

The term K_A/K_B is just the production ratio in sudden synthesis. To facilitate comparison of different nuclides, we express all abundances as "reduced" abundances relative to those in sudden synthesis:

$$\frac{X_t^A}{X^B}\cdot\frac{K_B}{K_A}=\frac{\tau}{T}\cdot e^{-\Delta t_{XY}/\tau}. \tag{27}$$

For a given T, these reduced abundances are a function of τ and Δt only. If Δt is fixed, the reduced abundances lie on an isochrone. Three such isochrones for $T=10^{10}$ y and $\Delta t=0$, 50, and 100 Myr are shown in Fig. 14. The point for Pd^{107} was calculated from Murthy's (1961b) data for Canyon Diablo, setting $Pd/Ag\approx 100$ (Goles 1961), $K_{107}/K_{105}=0.87$ (Burbidge *et al.* 1957) and assuming terrestrial silver to represent primeval Canyon Diablo silver. The points for I^{129} (Richardton and Indarch) were taken from Table XVI, and the point for Pb^{205}, from Table XVII. The point for Pu^{244} was also included, although it refers to the Earth rather than to the meteorites.

The points for the three longer-lived extinct nuclides agree tolerably well with the 100 Myr isochrone. Had the same Δt applied to Pd^{107}, its reduced abundance would have been 4×10^{-8}. Actually, a value of $\sim 1.1\times 10^{-3}$ has been observed!

Clearly, no reasonable change in the parameters of this calculation can bridge the gap between these two values. One must conclude that a much greater amount of Pd^{107} was present in the meteorite parent bodies at the time of their solidification than would be expected for a decay interval the order of 10^8 yr. Indeed, the amount is quite comparable to the expected steady-state abundance in the Galaxy at $\Delta t=0$.

This paradox can be explained in either of two ways (Anders 1961). The I-Xe method actually measures the cooling of the meteorite parent bodies to some temperature the order of 300°K where xenon retention could begin, whereas the Pd-Ag method measures the cessation of melting in these bodies. Even for bodies as small as 100 km in radius, these times can be appreciable, as shown in Table XIV. The Pu-Xe method also measures cooling to low temperatures. Moreover, the only point available for this method refers to the Earth rather than to the meteorite parent bodies. The Pb-Tl method, on the other hand, dates the same solidification event as the Pd-Ag method; in fact, for the same meteorite, but the uncertainties pointed out in Sec. 6.3, combined with a liberal estimate of errors in the Pd^{107} point, might conceivably bring the two points into accord at some Δt near 10 or 20 Myr. The absolute value of this Δt is, of course, subject to the usual uncertainties in T and other parameters, and might be larger or smaller than the values indicated. This would imply a rather short time for accretion, melting, and solidification of the meteorite parent bodies, followed by a longer cooling time.

Alternatively, one may propose a nuclear process that manufactures Pd^{107} in preference to other extinct radionuclides. This process must have occurred very shortly before the formation of the meteorite parent bodies, for, otherwise, any enhancement in the yield of Pd^{107} would be offset by its relatively faster decay. These requirements point rather strongly to the occurrence of local nuclear processes before or during the formation of the solar system, possibly caused by the

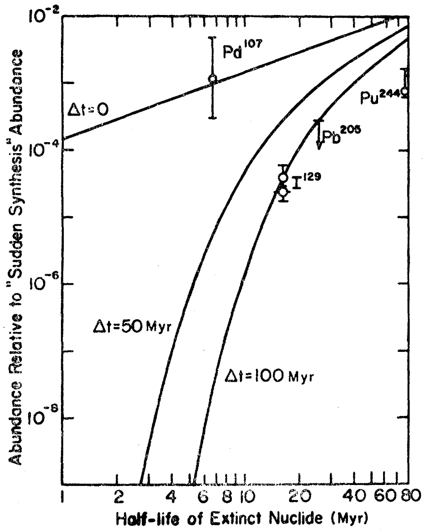

FIG. 14. Predicted and observed abundances of extinct radionuclides at various times (Δt) after the isolation of the solar system (Anders 1961). The experimental points are for the following objects: Pd^{107}, Canyon Diablo; I^{129}, Richardton and Indarch; Pb^{205}, Canyon Diablo; Pu^{244}, Earth. The high abundance of Pd^{107} implies either a long cooling time for the meteorite parent bodies, or preferential production of Pd^{107} during the formation of the solar system.

Table XIX. Noble gases in Pesyanoe meteorite. (Gas contents in 10^{-8} cc at STP.)

Object	He^4	Ne^{20}	Ar^{36}	$\frac{He^4}{He^3}$	$\frac{Ne^{20}}{Ne^{21}}$	$\frac{Ne^{20}}{Ne^{22}}$	$\frac{Ar^{36}}{Ar^{38}}$	$\frac{Ar^{40}}{Ar^{36}}$	Reference
Pesyanoe	7.3×10^5	2150	159	≥3600	114	11.3	5	28	b
Chondrites (typical)	1500	7	2	100	0.97	0.92	2	3000	c–e
Atmosphere[a]	25	86	159	8×10^5	354	10.3	5.35	295	

[a] Concentrations in the atmosphere are given in arbitrary units.
[b] Gerling and Levskii (1956).
[c] Eberhardt and Hess (1960).
[d] Eberhardt and Eberhardt (1961).
[e] Geiss and Hess (1958).

acceleration of charged particles during the dissipation of the Sun's magnetic field (Fowler 1960; Cameron 1960; Fowler, Greenstein, and Hoyle 1961).

6.5 General Isotopic Anomalies

Some additional light has been shed on the Ag^{107} problem by the discovery of "general" isotopic anomalies in meteorites. Reynolds (1960a) noted that, in addition to the anomaly at mass number 129, meteoritic and terrestrial xenon differed at other mass numbers as well (Table XV).

Differences at heavier mass numbers can probably be explained in terms of a fission contribution to terrestrial xenon (Sec. 6.2). For the lighter mass numbers, three different interpretations have been proposed. Goles and Anders (1961a) normalize the data to Xe^{130}, find an excess of the lighter isotopes in meteoritic xenon, and attribute this to charged-particle-induced, spallation reactions in the solar nebula. The difference between meteoritic and terrestrial xenon is attributed to the lower density of matter in the asteroidal belt, which may lead to higher levels of particle acceleration and spallation.

Cameron (1961) normalizes the data to Xe^{128}, and considers terrestrial xenon to be deficient in the light isotopes. He attributes this deficiency to the addition of solar xenon to the Earth's atmosphere, assuming that solar xenon was once irradiated by 100-ev neutrons during the deuterium-burning stage of the Sun's evolution. This irradiation would slightly deplete the isotopes 124, 126, 131, 134, and 136 while enhancing isotopes 128 and 130.

Krummenacher *et al.* (1961) normalize the data to Xe^{136} and conclude that the isotopic differences can be explained by: (a) addition of fission-product xenon to the meteorites and (b) strong mass fractionation of terrestrial xenon, leading to progressive depletion of the lighter isotopes.

All three hypotheses can be made to account in some *ad hoc* manner for the observation that meteoritic krypton shows much smaller anomalies, the order of 2% or less. On this basis alone, none can be disqualified, though the plausibility of these *ad hoc* assumptions varies. Arguments against mass fractionation in particular are given in Sec. 7. However, Umemoto (1962) has observed isotopic anomalies in meteoritic barium that parallel those of xenon very closely. Barium is a nonvolatile element, and, hence, neither the mechanism of Cameron nor that of Krummenacher *et al.* can be invoked to explain the anomalies without further *ad hoc* assumptions, since both depend on the fractionation or transport of gases. Thus, the present evidence seems to favor the mechanism of Goles and Anders (1961a) or, more generally, some type of nuclear process that affected meteoritic matter to a greater extent than terrestrial matter.

For a definitive picture, further data on other chemical elements are needed. Earlier workers failed to detect isotopic anomalies in many elements isolated from meteorites (Brown 1949; Schmitt, Mosen, Suffredini, Lasch, Sharp, and Olehy 1960; Reed, Kigoshi, and Turkevich 1960). But, since the anomalies appear to be of the order of 2% or less for nonvolatile elements, it seems likely that many anomalies were missed due to insufficient sensitivity of the analytical methods employed.

A detailed mechanism for the production of such anomalies was proposed by Fowler *et al.* (1961). They suggest that intense irradiation with 500-Mev protons and thermal neutrons took place at a time when most planetary and meteoritic matter was contained in meter-sized planetesimals. The dimensions of the planetesimals must have been such that, on the average, only some 10% of the material was strongly irradiated.

Undoubtedly, this picture will be refined and modified as further isotopic data become available. Already on the basis of the present, fragmentary data, a large number of possibilities has been ruled out conclusively. Each nuclear process produces its own characteristic abundance pattern, and it is thus possible, given a sufficiently detailed pattern, to deduce the processes that were responsible for it. It seems very likely that within a few years the nuclear events that preceded or accompanied the formation of the solar system will be known in greater detail than any other aspect of that era.

7. PRIMORDIAL NOBLE GASES

In 1956, Gerling and Levskii reported the discovery of unusually large amounts of noble gases in the aubrite Pesyanoe. Not only were the amounts of He and Ne much greater than in any meteorite studied up to that time, but the isotopic ratios, too, were radically

TABLE XX. Primordial noble gases in meteorites. [10^{-8} cc/g (at STP)].

	Class	He^4	Ne^{20}	Ar^{36}	$\frac{\mathrm{He}^4}{\mathrm{Ne}^{20}}$	$\frac{\mathrm{Ne}^{20}}{\mathrm{Ar}^{36}}$	$\log\frac{\mathrm{Ar}^{36}}{\mathrm{Si}}$	$\frac{\mathrm{He}^4}{\mathrm{He}^3}$	$\frac{\mathrm{Ne}^{20}}{\mathrm{Ne}^{22}}$	$\frac{\mathrm{Ar}^{36}}{\mathrm{Ar}^{38}}$	Reference
Nuovo Urei	U		<1.3	238		<0.0055	-7.80			5.36	a
Goalpara	U		<1.0	39		<0.026	-8.59			5.88	a
Lancé	Kc		3.3	174		0.019	-7.85			5.36	a
Felix	Kc		5.6	167		0.031	-7.88			5.40	a
Ivuna	K		20.9	96		0.22	-7.94		9.8	5.37	a
Abee	Cek		6.3	25		0.25	-8.75		9.3	5.50	b
Murray	K		53.8	137		0.39	-7.89		10.6	5.37	a
Atmosphere						0.52	-9.76		10.2	5.35	
Washington Co.	D_1	1500	36	...	420	...	...				c, d
Murray	K	1.3×10^4	63.2	76	210	0.83	-8.15		9.2	5.47	e
Mokoia	Kc		309	34	...	9.1	-8.56		13.2	5.39	a
Pantar (dark)	Ca	1.1×10^5	394	...	280	...	...	2900	12.4 ± 0.3	...	f
Pesyanoe	Au	7.3×10^5	2150	159	340	11.9	-8.06	≥3600	12.2	5.46	a, g
Kapoeta	Ho	1.37×10^5	2400	107	57	22.4	-8.23	3200	13.9	5.20	b
"Cosmos"		...	...	...	400	61.4	-0.9	...	23.0	7.87	h

[a] Stauffer (1961a).
[b] Zähringer and Gentner (1960).
[c] Schaeffer and Fisher (1959).
[d] Signer and Nier (1961).
[e] Reynolds (1960b).
[f] König, Keil, Hintenberger, Wlotzka, and Begemann (1961).
[g] Gerling and Levskii (1956).
[h] Suess and Urey (1956).

different (Table XIX). The preponderance of light noble gases and the low $\mathrm{Ar}^{40}/\mathrm{Ar}^{36}$ ratio rules out atmospheric contamination as a principal source of these gases. A cosmogenic origin was ruled out by the low He^3 content and by the neon isotopic ratios, since spallation would have produced all three neon isotopes in comparable amounts. Finally, no more than a small fraction of the He^4 could have arisen from the radioactive decay of U and Th in the meteorite.

It, therefore, seemed likely that the "excess" gases were largely primordial. For several years, Pesyanoe remained the only known case of its kind, until Schaeffer and Fisher (1959) found excess He^4 in the Washington County iron. Later, Zähringer and Gentner (1960) discovered primordial gases in the enstatite chondrite Abee and the calcium-rich achondrite Kapoeta, while Reynolds (1960b) observed them in the carbonaceous chondrite Murray. Stauffer (1961a) confirmed the results for Murray and Pesyanoe, and extended the measurements to four other carbonaceous chondrites and two ureilites. Finally, König, Keil, Hintenberger, Wlotzka, and Begemann (1961) discovered primordial gases in the ordinary chondrite Pantar. Thus, it seems that primordial noble gases occur among all major classes of stone meteorites (Table XX).[12]

7.1 Mechanism of Trapping

One of the most interesting questions raised by the presence of these gases is the nature of the trapping mechanism. In the absence of any clues to the distribution of the gases among the meteoritic minerals, Gerling and Levskii assumed that the gases were simply dissolved in the silicates at the time of solidification, and that the partition between the melt and the gas phase obeyed Henry's law. Taking the solubility of all noble gases at 1500°–2000°C as identical to the value for He in a gabbro-diabase melt, 2.84×10^{-3} cm^3/g, they calculated the partial pressures of He ($\geq$2200 mm), Ne ($\sim$9 mm), and Ar (0.7 mm) in the gas phase, and concluded that such high partial pressures of He could prevail only in the atmosphere of a large, planetary body.

This problem was reexamined by DuFresne and Anders (1961a), who investigated Pesyanoe without success for silicate minerals with specific gas-trapping structures (beryl, cordierite, etc.). They argued on steric grounds that the gases were most likely to be trapped in lattice defects and dislocations of a close-packed structure with large anion radius, e.g., MgS, CaS, or FeS. In fact, x-ray patterns of MgS and FeS from Pesyanoe revealed a considerable degree of disorder. To avoid the uncomfortable implications of a dense, planetary atmosphere, they suggested that the high partial pressures of noble gases required by Gerling and Levskii could also have arisen in a closed magma chamber. Although an asteroidal body is far too small to hold noble gases gravitationally, it could still retain them by another mechanism (DuFresne and Anders 1961a, b). Such a body, if accreted from a cold nebula of solar composition, would initially contain an appreciable endowment of noble gases. Internal heating would release these gases (and other volatiles, e.g., H_2O) from the center, and cause them to migrate outward. The surface of this body would be colder than 200°K, so that water vapor migrating toward the surface from below would condense and then freeze to a

---

[12] Geiss and Hess (1958) noted a slight excess of Ar^{36} in a number of ordinary chondrites ($\mathrm{Ar}^{36}/\mathrm{Ar}^{38}=1.18$ to 3.8, compared to the cosmogenic ratio of 0.7). It has not been established whether this excess is due to primordial gas, or to a slight amount of neutron capture in Cl^{35}, followed by β decay of Cl^{36} (Geiss 1957; Eberhardt, Geiss, and Lutz 1961).

permafrost layer. [Such a layer had been previously postulated for the Moon by Gold (1961)]. This permafrost layer might act as a barrier to the escape of gases, and might be aided in this by the sintered silicate layer at greater depths, postulated by Fish, Goles, and Anders (1960). In this manner, a temporary "internal atmosphere" might develop in the body.

Granted the existence of an internal reservoir of gas, another attractive possibility for the trapping of gas would be crystal growth by reaction of solid minerals with hot gases; such a process seems to be suggested by the close, physical association of MgS and $MgSiO_3$ (DuFresne and Anders 1961a). Fish *et al.* had previously inferred from the observed enrichment of sulfides in meteoritic veins, and other evidence, that hot, sulfur-containing gases might have been released explosively from the interior of the meteorite parent bodies. If this gas steam had also contained noble gases from the degassing of primordial matter in the interior, any sulfide crystals grown by vapor-solid reactions of the outflowing vapors would incorporate some fraction of the noble gases present.

Although the postulated enrichment of noble gases in the sulfide minerals of Pesyanoe has not yet been verified experimentally, this mechanism receives some support from the work of König *et al.*, who found that the primordial noble gases in the Pantar chondrite were distributed quite nonuniformly. This meteorite consists of light gray, centimeter-sized inclusions, surrounded by a dark gray matrix that has apparently undergone severe thermometamorphism. The dark fraction is ~100 and ~50 times richer in primordial He and Ne than the light fraction. This implies that the meteorite has experienced very little reheating since the incorporation of the noble gases, for otherwise the gases would have become uniformly distributed (in a closed system), or lost (in an open system). Much, if not all, of the gas is contained in the magnetic fraction of the meteorite (consisting of metal and the magnetic variety of FeS), and since the dark fraction owes its color to fine veins consisting for the most part of "newly-formed" FeS, the gases may well have been incorporated in this mineral by a process not unlike that suggested by DuFresne and Anders (1961a).

Some valuable information on the present location of the trapped gases comes from heating experiments (Zähringer and Gentner 1961; Jeffery and Reynolds 1961a, b; see also Sec. 6.1). These experiments show that primordial Ar^{36} and Xe^{132} in the Abee meteorite are held much more strongly than radiogenic Ar^{40} (Fig. 12). At least in this meteorite, there is no question that the primordial gases are not simply adsorbed on surfaces, but are securely held in a retentive crystal lattice. Whether the gases are contained in one or several minerals cannot be determined at this time. Diffusion studies and measurements on separated mineral fractions may provide an answer to this question.

Note added in proof. König and Wlotzka (1962) were able to show that the primordial noble gases were indeed contained mainly in newly formed troilite and finely divided metal.

7.2 Elemental and Isotopic Fractionation of Primordial Gases

For a reconstruction of the meteorites' history, no other property of the noble gases has been as helpful as their volatility. It was shown in Sec. 5.3 and 6.1 that the retention or loss of radiogenic Ar^{40}, He^4, and Xe^{129} provided much valuable information on the cooling time, size, and breakup time of the meteorite parent bodies. All these dating methods depend on the production and retention of radiogenic gases, and the earliest datable event, therefore, is the end of the high-temperature era in the meteorite parent bodies, any radiogenic noble gases generated up to that time having been lost.

In contrast, the primordial noble gases were presumably incorporated in the very first solid matter that eventually accreted to the meteorite parent bodies. Each subsequent stage in the evolution of this matter offered some opportunities for elemental and isotopic fractionations of these gases. Though the present data are far too incomplete to permit a detailed reconstruction of this history, some tentative conclusions can nevertheless be drawn.

Stauffer (1961a) has pointed out several interesting trends in the data of Table XX. Comparison of the meteoritic Ar^{36}/Si ratios with the "cosmic" abundances of Suess and Urey (1956) shows that the meteorites have retained only about 10^{-7} of their original complement of primordial Ar^{36}. But, in spite of this large fractionation factor, the amounts retained are constant within a factor of 10 for all samples and within a factor of 5 for the carbonaceous chondrites. The Earth has retained only about 10^{-9} of its original complement.

In contrast, the retention of primordial neon, as shown by the spread in the Ne^{20}/Ar^{36} ratios, has been much more variable. Stauffer found for the ornansite Lancé that the D/a^2 value at 950°K for primordial $Ne^{21}(1.8\times10^{-6}\ \text{sec}^{-1})$ was greater than that for primordial $Ar^{36}(5.6\times10^{-7})$, though not as great as that for radiogenic $Ar^{40}(5.3\times10^{-6})$. Losses of the latter are indicated by the short K-Ar ages of many meteorites in Table XX. It, therefore, seems likely that some neon was lost as well at a relatively late stage in the development of the meteorites. The magnitude of the losses would in each case depend on the thermal history of the meteorite (Sec. 5.3), so that the variability of the present Ne/Ar ratios does not rule out the possibility of a constant ratio in the trapped primordial gas.

Helium should have been lost even more easily than neon, and the closest approximation to the initial composition of the primordial gases is, therefore, given by the 4 meteorites containing primordial He^4. For ease of comparison, the depletion factors (defined as the ratio of the observed abundance to the "cosmic" abundance) have been plotted against mass number (Fig. 15). Data for the Earth and the enstatite chondrite Abee have also been included.

In the case of the Earth, Suess (1949) drew attention to the fact that Kr and Xe had virtually the same depletion factor of $\sim 10^{-7}$, whereas Ar and particularly Ne were depleted by larger factors. The data could be represented by the equation[11]

$$-\log \frac{N_{\text{ter}}}{N_{\cos}} = 10e^{-0.045M/m_1} + 7.1, \tag{28}$$

which is identical in form to the equation for mass fractionation during escape of a gas from a gravitational field,

$$-\ln(N_t/N_0) = tAe^{-Mgr/RT}, \tag{29}$$

where N_{ter} = terrestrial abundance, $N_{\cos}$ = "cosmic" abundance; N_0 = initial abundance; N_t = abundance at time t; M = molecular weight; m_1 = atomic-weight unit; A = constant; g = gravitational acceleration in escape layer; r = distance of escape layer from center of gravity; R = gas constant; and T = temperature.

For gases of high M, the fractionation factor in Eq. (28) approaches the asymptotic value of $10^{-7.1}$. Suess, therefore, suggested that the gas loss from the earth took place in two stages: first, a general depletion (without mass fractionation) to $10^{-7.1}$ the initial value, followed by a second stage involving mass fractionation. He pointed out, however, that the small value of the exponent $Mgr/RT = 0.045\ M/m_1$ presented some difficulties. With the present value for the Earth's gravitational acceleration, no escape of neon, let alone heavier gases, is possible even assuming the extreme conditions of $T = 3000$°K and $r = 15\,000$ km. Consequently, Suess argued that the escape occurred either during a planetesimal stage preceding the formation of the Earth, or during an era when the Earth's period of rotation was so short that centrifugal force largely offset the gravitational force in the upper atmosphere.

This mechanism may or may not be valid for the Earth, but it is evident from Fig. 15 that at least the data for meteorites 2 to 5 do not fit an exponential fractionation function of the form of Eq. (28). Any such function with an exponent large enough to reproduce the observed fractionation of neon relative to xenon must, by necessity, predict a very large depletion of helium. For example, Eq. (28) predicts a depletion of He relative to Ne by a factor of 2×10^4. The observed depletion in meteorites amounts to less than a factor of two in 3 out of 4 cases, and only a factor of seven in the one remaining case![12]

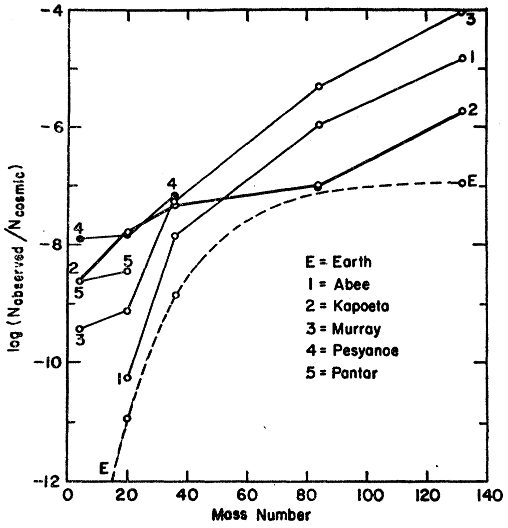

FIG. 15. Depletion factors (ratio of observed to "cosmic" abundance) for noble gases in meteorites (for references see Table XX). Data for the Earth are shown for comparison. The broken line indicates the Suess (1949) fractionation function, with the constant changed from 7.1 to 6.9.

If the fractionation of noble gases in meteorites were gravitational, He should be depleted far more strongly than Ne. This is not borne out by the data.

The isotopic ratios also do not agree with the expected values for gravitational fractionation. Geiss (1957) has used Eq. (28) to calculate the neon and argon isotopic composition in primordial matter from the terrestrial isotopic ratios. With the exception of Goalpara, the observed meteoritic Ar^{36}/Ar^{38} ratios in Table XX are exceedingly close to the atmospheric value of 5.35, much below the calculated primordial value of 7.87. In the case of neon, the agreement with the calculated primordial value is not much better.

Thus it seems unlikely that the meteoritic noble gases were fractionated gravitationally. For the terrestrial gases, a better case can be made, although it is not certain what proportion of these gases is actually of terrestrial origin. Urey (1952b) suggested that an appreciable addition to the Earth's noble-gas inventory might have been made by cometary material. Assuming a total accretion of 4g of cometary matter per cm² of the Earth's surface, he concluded that this source could account for only 1/50 of the Earth's xenon and 1/10 of the krypton. However, even at this low accretion rate (which is some 10^{-2} times smaller than recent estimates of micrometeorite influx rates), all of the Earth's neon could have been derived from comets. If the rate of influx of cometary matter had been greater in the past than in recent times, much of the Earth's noble gas inventory might have been derived from this source. Urey (1952c) suggested that the heavier noble gases

---

[11] Better agreement with the Suess and Urey (1956) abundances is obtained by changing the constant in Eq. (28) from 7.1 to 6.9. This change has been made in Fig. 15.

[12] Suess (1962) has pointed out that the meteoritic data in Fig. 15 could still be explained in terms of his model (loss of all but $\sim 10^{-4}$ of the initial gas by hydrodynamic flow, followed by gravitational fractionation of the remainder), if a small amount of unfractionated gas were retained or added. This suggestion has merit, although an *ad hoc* mechanism of retention or addition is required.

might be enriched in comets, owing to their greater condensability. The observed elemental fractionations would then reflect condensation conditions in the outer portions of the solar nebula, rather than gravitational fractionation of the earth's initial noble gas endowment. Urey points out that Suess' mechanism would lead to a large fractionation of the neon isotopes, whereas his would not. The observed slight differences between terrestrial and meteoritic neon are not in contradiction with Urey's hypothesis, if it is remembered that almost any nuclear process will lower the Ne^{20}/Ne^{22} ratio. Possibly the cometary neon experienced a slightly different nuclear history before being incorporated in the Earth.

Another perturbation to be considered is the influx of neutron-irradiated noble gases (albeit Kr and Xe only) from the Sun as suggested by Cameron (1961).

In view of these factors, one should bear in mind that the terrestrial noble-gas abundances, though consistent with gravitational fractionation, do not necessarily imply that the fractionation actually proceeded by this mechanism. It is not altogether impossible that the same process (or combination of processes) which fractionated the noble gases in the meteorites was also responsible for the fractionations in the Earth, and that gravitational fractionation was of little importance in either case.

Even though strongly mass-dependent processes, such as gravitational fractionation, are ruled out as the principal mechanism for the meteorites, since they would lead to a much smaller retention of He than observed, there is little doubt that the fractionation process was slightly mass-dependent, as shown by the progressively better retention of the heavier noble gases. One must, therefore, examine other processes that can account for the data.

(1) Diffusion, too, is a mass-dependent process, though to a lesser and much more variable degree. The diffusive loss of a gas depends on the parameter D/a^2 (Sec. 5.3), where

$$D = D_0 e^{-Q/RT} \tag{30}$$

(Q=activation energy; D_1=a constant). Both Q and D_0 vary from one mineral and one gas to another, the heavier gases and the more-densely packed minerals generally having larger values of Q. It is seen from Eq. (30) that the fractionation between two gases will be strongly temperature-dependent, any differences in D being accentuated at low temperatures. The diffusion mechanism can thus be invoked to account for almost any observed fractionation, large or small, particularly if the gases are unevenly distributed among several minerals differing in retentivity and grain size. Such diffusion losses could have occurred not only during or after the cooling of the meteorite parent bodies (Sec. 5.3). but also during the entire period from the formation of solid grains in the solar nebula to the end of the high-temperature era in the parent bodies.

(2) Adsorption was rejected by Suess (1949) as not capable of giving the large, observed fractionation between Ne and the heavier gases. However, at low temperatures, noble gases differ sufficiently in their adsorption behavior to permit their separation in the laboratory. At least the heavier gases could have been retained in this manner, and the poor adsorbability of He and Ne may well explain the greater (and nearly equal) depletion of these two elements.

(3) Differential solubility was also dismissed by Suess as too inefficient a mechanism to give fractionation factors as large as 10^4. This argument may still be valid, though it is perhaps based on too limited a body of experimental evidence. Preferential trapping of the heavy noble gases as hydrates has been considered by Miller (1961), and the solubility in solids may be more selective than the solubility in liquids discussed by Suess.

Quite possibly, more than one process was responsible for the fractionation. Tracing the history of a gas molecule from the moment of trapping in an interstellar (or nebular) grain to its final release in an induction furnace, one cannot help but realize the great variety of physical conditions it experienced. Particularly during its residence in the meteorite parent body, temperature gradients must have been great enough to permit extensive fractionation by desorption, adsorption, diffusion, and solution. The lack of fractionation between He and Ne is all the more remarkable in view of these diverse opportunities. These and other observations on primordial gases may eventually be of great value in delineating the earliest history of the solar system, complementing the nuclear information discussed in Sec. 6.

ACKNOWLEDGMENTS

Part of this paper was written at the Theoretical Division, Goddard Space Flight Center, National Aeronautics and Space Administration, Greenbelt, Maryland. The author is indebted to the National Academy of Sciences for a Senior Postdoctoral Resident Research Associateship during his stay at Goddard, and to the U. S. Atomic Energy Commission for subsequent support at The University of Chicago.

I also want to thank friends and colleagues who contributed advice, criticism, or unpublished data, in particular, Dr. E. L. Fireman, Dr. D. E. Fisher, Dr. P. W. Gast, Dr. G. G. Goles, Dr. J. A. O'Keefe, Dr. E. L. Segel, Dr. S. P. Shen, and Dr. H. Wänke.

BIBLIOGRAPHY

D. W. Allan and J. A. Jacobs (1956), Geochim. et Cosmochim. Acta **9**, 256 (1956).

E. Anders (1961), Z. Naturforsch. **16a**, 520 (1961).

E. Anders and G. G. Goles (1961), J. Chem. Educ. **38**, 58 (1961).

E. Anders and C. M. Stevens (1960), J. Geophys. Research **65**, 3043 (1960).

J. R. Arnold, M. Honda, and D. Lal (1961), J. Geophys. Research **66**, 3519 (1961).

W. J. Arrol, R. B. Jacobi, and F. A. Paneth (1942), Nature **149**, 245 (1942).

A. Bainbridge, H. E. Suess, and H. Wänke (1961), Geochim. et Cosmochin. Acta (to be published).

G. L. Bate, J. R. Huizenga, and H. A. Potratz (1959), Geochim. et Cosmochim. Acta **16**, 88 (1959).

C. A. Bauer (1947), Phys. Rev. **72**, 354 (1947).

C. A. Bauer (1948), Phys. Rev. **74**, 225 (1948).

C. A. Bauer (1960), Astron. J. **65**, 340 (1960).

C. A. Bauer (1961), (private communication).

F. Begemann, J. Geiss, and D. C. Hess (1957), Phys. Rev. **107**, 540 (1957).

F. Begemann, P. Eberhardt, and D. C. Hess (1959), Z. Naturforsch. **14a**, 500 (1959).

R. L. Bjork (1961), J. Geophys. Research **66**, 3379 (1961).

H. Brown (1947), Phys. Rev. **72**, 348 (1947).

H. Brown (1949), Revs. Modern Phys. **21**, 625 (1949).

H. S. Brown and E. Goldberg (1949), Phys. Rev. **76**, 1260 (1949).

J. D. Buddhue (1941), Popular Astron. **60**, 390 (1941).

E. M. Burbidge, G. R. Burbidge, W. A. Fowler, and F. Hoyle (1957), Revs. Modern Phys. **29**, 548 (1957).

E. S. Burkser, F. I. Kotlovskaja, and B. B. Zaidis, (1958), Geol. Zhur. Akad. Nauk. Ukrain. SSR Inst. Geol. Nauk **18**, 90 (1958).

A. G. W. Cameron (1959), Astrophys. J. **129**, 676 (1959).

A. G. W. Cameron (1960), *Proceedings of the Highland Park Conference on Interplanetary Matter, June 20–22* (National Academy of Science-National Research Council Publication No. 845).

A. G. W. Cameron, (1961) (to be published).

V. V. Cherdyntsev and K. K. Abdulgafarov (1956), Doklady Akad. Nauk S.S.S.R. **106**, 311 (1956).

V. V. Cherdyntsev (1961), *Abundance of Chemical Elements* (University of Chicago Press, Chicago, Illinois, 1961).

T. J. Chow and C. C. Patterson (1959), Geochim. et Cosmochim. Acta **17**, 21 (1959).

W. B. Clarke and H. G. Thode (1961), J. Geophys. Research **66**, 3578 (1961).

J. C. Dalton, J. Golden, G. R. Martin, E. R. Mercer, and S. J. Thomson (1953), Geochim. et Cosmochim. Acta **3**, 272 (1953).

J. C. Dalton, F. A. Paneth, P. Reasbeck, S. J. Thomson, and K. I. Mayne (1953), Nature **172**, 1168 (1953).

J. C. Dalton and S. J. Thomson (1954), Geochim. et Cosmochim. Acta **5**, 74 (1954).

G. L. Davis (1950), Am. J. Sci. **248**, 107 (1950).

R. Davis (1959) (unpublished).

S. Deutsch, F. G. Houtermans, and E. E. Picciotto (1956), Geochim. et Cosmochim. Acta **10**, 166 (1956).

L. Drummeter and M. Schach (1958), in *Annals of the International Geophysical Year*, edited by L. D. Berkner (Pergamon Press, New York, 1958), Vol. 6, p. 304.

E. R. DuFresne and E. Anders (1961a), Geochim. et Cosmochim. Acta **26**, 251 (1962).

E. R. DuFresne and E. Anders (1961b) Geochim. et Cosmochim. Acta (to be published).

P. Eberhardt and A. Eberhardt (1961), Z. Naturforsch. **16a**, 236 (1961).

P. Eberhardt and D. C. Hess (1960), Astrophys. J. **131**, 38 (1960).

P. Eberhardt and J. Geiss (1960a), Lecture at the Varenna Summer School on Nuclear Geology, September 1960, (to be published).

P. Eberhardt and J. Geiss (1960b), Z. Naturforsch. **15a**, 547 (1960).

P. Eberhardt J. Geiss, and F. G. Houtermans (1955), Z. Physik **141**, 91 (1955).

P. Eberhardt, J. Geiss, and H. Lutz (1961), Helv. Phys. Acta **34**, 460 (1961).

K. H. Ebert, F. Hernegger, H. König, and H. Wänke (1959), Geochim. et Cosmochim. Acta **17**, 349 (1959).

K. H. Ebert and H. Wänke (1957), Z. Naturforsch. **12a**, 766 (1957).

G. Edwards (1955), Geochim. et Cosmochim. Acta **8**, 285 (1957).

G. Edwards and H. C. Urey (1955), Geochim. et Cosmochim. Acta **7**, 154 (1955).

H. Fechtig, W. Gentner, and G. Kistner (1960), Geochim. et Cosmochim. Acta **18**, 72 (1960).

E. L. Fireman (1958a), Nature **181**, 1613 (1958).

E. L. Fireman (1958b), Nature **181**, 1725 (1958).

E. L. Fireman (1959) Planetary, Space Sci. **1**, 66 (1959).

E. L. Fireman and J. DeFelice (1960a), Geochim. et Cosmochim. Acta **18**, 183 (1960a).

E. L. Fireman and J. DeFelice (1960b), J. Geophys. Research **65**, 3035 (1960).

E. L. Fireman and J. DeFelice (1961), J. Geophys. Research **66**, 3547 (1961).

E. L. Fireman and D. E. Fisher (1961), Nature **192**, 644 (1961).

R. A. Fish, G. G. Goles, and E. Anders (1960), Astrophys. J. **132**, 243 (1960).

D. E. Fisher (1961a), Nature **190**, 244–5 (1961).

D. E. Fisher (1961b), Nature **190**, 225–7 (1961).

D. E. Fisher (1961c), J. Geophys. Research **66**, 1509 (1961).

D. E. Fisher and O. A. Schaeffer (1960), Geochim. et Cosmochim. Acta **20**, 5 (1960).

K. F. Flynn and L. E. Glendenin (1959), Phys. Rev. **116**, 744 (1959).

R. E. Folinsbee, J. I. Lipson, and J. H. Reynolds (1956), Geochim. et Cosmochim. Acta **10**, 60 (1956).

W. A. Fowler (1960) (private communication).

W. A. Fowler, J. P. Greenstein, and F. Hoyle (1961) (to be published).

P. W. Gast (1960a), Geochim. et Cosmochim. Acta **19**, 1 (1960).

P. W. Gast (1960b) *Proceedings of the Highland Park Conference on Nuclear Geology*, National Academy of Science—National Research Council, Publication No. 845. Washington, D. C.

P. W. Gast (1960c), J. Geophys. Research **65**, 1287 (1960).

P. W. Gast (1961) (to be published).

J. Geiss and D. C. Hess (1958), Astrophys. J. **127**, 224 (1958).

J. Geiss, B. Hirt, and H. Oeschger (1960), Helv. Phys. Acta **33**, 590 (1960).

J. Geiss and H. Oeschger (1960) *Proceedings of the First International Space Science Symposium* (North-Holland Publishing Company, Amsterdam, 1960), p. 1071.

J. Geiss, H. Oeschger, and P. Signer (1960), Z. Naturforsch. **15a**, 1016 (1960).

E. K. Gerling and L. K. Levskii (1956), Doklady Akad. Nauk S.S.S.R. **110**, 750 (1956).

E. K. Gerling and T. G. Pavlova (1951), Doklady Akad. Nauk S.S.S.R. **77**, 85 (1951).

E. K. Gerling and K. G. Rik (1955), Doklady Akad. Nauk S.S.S.R. **101**, 433 (1955).

K. Goebel and P. Schmidlin (1959), Geochim. et Cosmochim. Acta **17**, 342 (1959).

K. Goebel, P. Schmidlin, and J. Zähringer (1959), Z. Naturforsch. **15a**, 996 (1959).

P. S. Goel (1960) (unpublished); see also *Researches in Meteorites*, edited by Carleton Moore (John Wiley & Sons, Inc., New York, 1962).

T. Gold (1961), paper presented at meeting of American Geophysical Union, April 19–22, Washington, D. C.

E. Goldberg, A. Uchiyama, and H. Brown (1951), Geochim. et Cosmochim. Acta **2**, 1 (1951).

V. M. Goldschmidt (1954), *Geochemistry* (Oxford University Press, New York, 1954).

G. G. Goles (1961) (private communication).

G. G. Goles and E. Anders (1960), J. Geophys. Research **65**, 4181 (1960).

G. G. Goles and E. Anders (1961a), J. Geophys. Research **66**, 889 (1961).

G. G. Goles and E. Anders (1961b) J. Geophys. Research **66**, 3075 (1961).

G. G. Goles and E. Anders (1961c), Geochim. et Cosmochim. Acta (to be published).

G. G. Goles and R. A. Fish (1961) (to be published).

G. G. Goles, R. A. Fish, and E. Anders (1960), Geochim. et Cosmochim. Acta **19**, 177 (1960).

I. Groeneveld and G. P. Kuiper (1954), Astrophys. J. **120**, 529 (1954).

H. Hamaguchi, G. W. Reed, and A. Turkevich (1957), Geochim. et Cosmochim. Acta **12**, 337 (1957).

E. P. Henderson and S. H. Perry (1958), Proc. U. S. Natl. Museum **107**, 339 (1958).

W. Herr, B. Hirt, and W. Hoffmeister (1961) (to be published).

W. Herr, W. Hoffmeister, B. Hirt, J. Geiss, and F. G. Houtermans (1961), Z. Naturforsch. **16a**, 1053 (1961).

W. Herr, W. Hoffmeister, J. Langhoff, J. Geiss, B. Hirt, and F. G. Houtermans (1960), Proceedings of the Conference on the Use of Radioisotopes in Phys. Sci. Industry, Copenhagen (to be published).

W. Herr and E. Merz (1958), Z. Naturforsch. **13a**, 231 (1958).

L. F. Herzog and W. H. Pinson, Jr. (1956), Am. J. Sci. **254**, 555 (1956).

D. C. Hess and R. R. Marshall (1960), Geochim. et Cosmochim. Acta **20**, 284 (1960).

D. C. Hess, R. R. Marshall, and H. C. Urey (1957), Science **126**, 1291 (1957).

M. H. Hey (1953), *Catalogue of Meteorites*, edited by G. T. Prior (British Museum, London, 1953), 2nd Ed., pp. 432.

D. Heymann and O. A. Schaeffer (1961), Paper presented at meeting of the American Geophysics Union, April 18–21, 1961, Washington, D. C. (unpublished).

H. Hintenberger, H. König, and H. Wänke (1961), Z. Naturforsch. (to be published).

J. H. Hoffman and A. O. Nier (1958), Phys. Rev. **112**, 2112 (1958).

J. H. Hoffman and A. O. Nier (1959), Geochim. et Cosmochim. Acta **17**, 32 (1959).

J. H. Hoffman and A. O. Nier (1960), J. Geophys. Research **65**, 1063 (1960).

M. Honda (1959), Geochim. et Cosmochim. Acta **17**, 148 (1959).

M. Honda and J. R. Arnold (1961), Geochim. et Cosmochim. Acta **23**, 219 (1961).

M. Honda, J. P. Shedlovsky, and J. R. Arnold (1961), Geochim. et Cosmochim. Acta **22**, 133 (1961).

M. Honda, S. Umemoto, and J. R. Arnold (1961), J. Geophys. Research **66**, 3547 (1961).

P. M. Jeffery and J. H. Reynolds (1961a), Z. Naturforsch. **16a**, 431 (1961).

P. M. Jeffery and J. H. Reynolds (1961b), J. Geophys. Research **66**, 3582 (1961).

S. Katcoff, O. A. Schaeffer, and J. M. Hastings (1951), Phys. Rev. **82**, 688 (1951).

V. G. Khlopin and E. K. Gerling (1948), Doklady Akad. Nauk S.S.S.R. **61**, 297 (1948).

T. P. Kohman (1953), *Nuclear Processes in Geological Settings* (National Academy of Sciences—National Research Council, Washington, D. C., 1953).

T. P. Kohman (1956), Proc. N. Y. Acad. Sci. **62**, 503 (1956).

T. P. Kohman (1961), J. Chem. Educ. **38**, 73 (1961).

T. P. Kohman and P. S. Goel (1961), Paper presented at Meteorite Symposium at Arizona State University, March 10, 1961, Temple, Arizona. Published in *Researches in Meteorites*, edited by Carleton Moore (John Wiley & Sons, Inc., New York, 1962).

H. König, K. Keil, H. Hintenberger, F. Wlotzka, and F. Begemann (1961), Z. Naturforsch. **16a**, 1124 (1961).

H. König and H. Wänke (1959), Geochim. et Cosmochim. Acta **17**, 350 (1959).

H. König, H. Wänke, and K. I. Mayne (1959), Geochim. et Cosmochim. Acta **17**, 339 (1959).

H. König and F. Wlotzka (1962), Z. Naturforsch. **17a** (to be published).

D. Krummenacher, C. M. Merrihue, R. O. Pepin, and J. H. Reynolds (1961), Geochim. et Cosmochim. Acta **26**, 231 (1962).

D. Krummenacher (1961), Helv. Chim. Acta **44**, 1054 (1961).

G. P. Kuiper (1950), Astron. J. **55**, 164 (1950).

P. K. Kuroda (1960), Nature **187**, 36 (1960).

P. K. Kuroda (1961), Geochim. et Cosmochim. Acta **24**, 40 (1961).

M. E. Lipschutz and E. Anders (1961), Geochim et Cosmochim. Acta **24**, 83 (1961).

J. F. Lovering, W. Nichiporuk, A. Chodos, and H. Brown (1957), Geochim. et Cosmochim. Acta **11**, 263 (1957).

J. F. Lovering, L. G. Parry, and J. C. Jaeger (1960), Geochim. et Cosmochim. Acta **19**, 156 (1960).

G. J. F. MacDonald (1959), J. Geophys. Research **64**, 1967 (1959).

R. E. Maringer and G. K. Manning (1960), Geochim. et Cosmochim. Acta **18**, 157 (1960).

R. R. Marshall (1959), Nature **184**, 117 (1959).

R. R. Marshall (1960), Nature **185**, 89 (1960).

R. R. Marshall (1962), Geochim. et Cosmochim. Acta (to be published).

R. R. Marshall and D. C. Hess (1961), Geochim. et Cosmochim. Acta **21**, 161 (1961).

G. R. Martin (1953), Geochim. et Cosmochim. Acta **3**, 288 (1953).

S. L. Miller (1961), Proc. Natl. Acad. Sci. U. S. (1961).

J. M. Miller and J. Hudis (1959), Ann. Rev. Nuclear Sci. **9**, 159 (1959).

V. R. Murthy (1960), Phys. Rev. Letters **5**, 539 (1960).

V. R. Murthy (1961a) (to be published).

V. R. Murthy (1961b) (to be published).

V. R. Murthy and C. C. Patterson (1961) (to be published).

V. R. Murthy and H. C. Urey (1962), Astrophys. J. **135**, 626 (1962).

H. H. Nininger (1956), *Arizona's Meteorite Crater* (American Meteorite Museum, Sedona, Arizona, 1956).

I. Noddack and W. Noddack (1934), Svensk. Kem. Tidskr. **46**, 173 (1934).

E. J. Öpik (1951), Proc. Roy. Irish Acad. **54**, 165 (1951).

E. J. Öpik (1958), Irish Astronom. J. **5**, 14 (1958).

E. J. Öpik and S. F. Singer, Trans. Am. Geophys. Union **38**, 566 (1957).

F. A. Paneth, H. Gehlen, and P. L. Guenther (1928), Z. Elektrochem. **34**, 645 (1928).

F. A. Paneth, P. Reasbeck, and K. I. Mayne (1952), Geochim. et Cosmochim. Acta **2**, 300 (1952).

C. C. Patterson (1955), *Nuclear Processes in Geologic Settings.* (Publication No. 400, National Academy Science—National Research Council).

C. C. Patterson (1956), Geochim. et Cosmochim. Acta **10**, 230 (1956).

J. H. Peebles and R. Dicke (1962) (to be published).

J. H. Piddington and H. C. Minnett (1949), Aust. J. Sci. Research **A2**, 63 (1949).

S. Piotrowski (1953) Acta Astronom. **5**, 115 (1953).

P. Reasbeck and K. I. Mayne (1955), Nature **176**, 733 (1955).

G. W. Reed (1961) (private communication).

G. W. Reed, H. Hamaguchi, and A. Turkevich (1958), Geochim. et Cosmochim. Acta **13**, 248 (1958).

G. W. Reed, K. Kigoshi, and A. Turkevich (1960), Geochim. et Cosmochim. Acta **20**, 122 (1960).

G. W. Reed and A. Turkevich (1955), Nature **176**, 794 (1955).

G. W. Reed and A. Turkevich (1957), Nature **180**, 594 (1957).

J. H. Reynolds (1960a), Phys. Rev. Letters **4**, 8 (1960).

J. H. Reynolds (1960b), Phys. Rev. Letters **4**, 351 (1960).

J. H. Reynolds (1960c), J. Geophys. Research **65**, 3843 (1960).

J. H. Reynolds (1960d), Z. Naturforsch. **15a**, 1112 (1960).

J. H. Reynolds and J. I. Lipson (1957), Geochim. et Cosmochim. Acta **12**, 330 (1957).

J. H. Reynolds, C. M. Merrihue, and R. O. Pepin (1962), paper presented at meeting of American Physical Society, New York, January 24–27, 1962.

D. B. Rosenblatt (1953), Phys. Rev. **91**, 1474 (1953).

S. G. Rudstam (1955), Phil. Mag. **46**, 344 (1955).

O. A. Schaeffer (1960), Nature **188**, 1102 (1960).

O. A. Schaeffer and D. E. Fisher (1959), Nature **183**, 660 (1959).

O. A. Schaeffer and D. E. Fisher (1960), Nature **186**, 1040 (1960).

O. A. Schaeffer and J. Zähringer (1959), Phys. Rev. **113**, 674 (1959).

R. A. Schmitt, A. W. Mosen, C. S. Suffredini, J. E. Lasch, R. A. Sharp, and D. A. Olehy (1960), Nature **186**, 863 (1960).

E. Schumacher (1956), Z. Naturforsch. **11a**, 206 (1956).

E. M. Shoemaker (1960), International Geological Congress XXI Session, pt. XVIII, p. 418.

P. Signer (1960), Z. Naturforsch. **15a**, 748 (1960).

P. Signer (1961a), Paper presented at meeting of American Geophysical Union, April 18–22, Washington, D. C. (unpublished).

P. Signer (1961b) (private communication).

P. Signer and A. O. Nier (1960), J. Geophys. Research **65**, 2947 (1960).

P. Singer and A. O. Nier (1961) in *Researches in Meteorites*, edited by Carleton Moore (John Wiley & Sons, Inc., New York, 1962).

S. F. Singer (1952), Nature **170**, 728 (1952).

S. F. Singer, Astrophys. J. **119**, 291 (1954).

S. F. Singer, Nuovo cimento **8**, 539 (1958).

S. F. Singer, Geochim. et Cosmochim. Acta **19**, 216 (1960).

S. F. Singer, Irish Astron. J. **4**, 165 (1957).

E. L. Sprenkel (1959), "Cosmic-Ray Produced Chlorine-36 in Iron Meteorites." PhD. thesis, University of Rochester, (unpublished).

I. E. Starik, M. M. Shats, and E. V. Sobotovich (1958), Doklady Akad. Nauk S.S.S.R. **123**, 424 (1958).

I. E. Starik, E. V. Sobotovich, G. P. Lovtsyus, M. M. Shats, and A. V. Lovtsyus (1959), Doklady Akad. Nauk S.S.S.R. **128**, 688 (1959).

I. E. Starik, E. V. Sobotovich, G. P. Lovtsyus M. M. Shats, and A. V. Lovtsyus (1960), Doklady Akad. Nauk S.S.S.R. **134**, 555 (1960).

H. Stauffer (1960) (unpublished).

H. Stauffer (1961a), Geochim. et Cosmochim. Acta **24**, 70 (1961).

H. Stauffer (1961b), Paper presented at meeting of American Geo-

H. Stauffer (1961c), J. Geophys. Research **66**, 1512 (1961).

physical Union, April 18–22, Washington, D. C. (unpublished).

H. Stauffer and M. Honda (1960), J. Geophys. Research **66**, 3584 (1961).

R. W. Stoenner, O. A. Schaeffer, and R. Davis, Jr. (1960) J. Geophys. Research **65**, 3025 (1960).

R. W. Stoenner and J. Zähringer (1958), Geochim. et Cosmochim. Acta **15**, 40 (1958).

H. E. Suess (1949), J. Geol. **57**, 600 (1949).

H. E. Suess and H. C. Urey (1956), Revs. Modern Phys. **28**, 53 (1956).

H. E. Suess and H. Wänke (1961), Geochim. et Cosmochim. Acta (to be published).

H. E. Suess (1962) (private communication).

S. R. Taylor and K. S. Heier (1958), Nature **182**, 202 (1958).

R. N. Thomas and F. L. Whipple (1951), Astrophys. J. **114**, 448 (1951).

S. J. Thomson and K. I. Mayne (1955), Geochim. et Cosmochim. Acta **7**, 169 (1955).

A. V. Trofimov and K. G. Rik (1955), Doklady Akad. Nauk S.S.S.R. **107**, 911 (1955).

A. Turkevich (1961) (private communication).

S. Umemoto (1962), J. Geophys. Research **67**, 375 (1962).

H. C. Urey (1952), Phys. Rev. **88**, 248 (1952).

H. C. Urey (1952b), *The Planets* (Yale University Press, New Haven, Conn., 1952).

H. C. Urey (1952c), L. Farkas Memorial Volume, Res. Council Israel Spec. Publ. #1, Jerusalem.

H. C. Urey (1955), Proc. Nat. Acad. Sci. U. S. **41**, 127 (1955).

H. C. Urey (1956a), Proc. Nat. Acad. Sci. U. S. **42**, 889 (1956).

H. C. Urey (1956b), Astrophys. J. **124**, 623 (1956).

H. C. Urey (1957a), 41st Guthrie Lecture, Yearbook Phys. Soc. **14** (1957).

H. C. Urey (1957b), Progress Phys. and Chem. Earth **2**, 46 (1957).

H. C. Urey (1958), Proc. Chem. Soc. **67**, (1958).

H. C. Urey (1959), J. Geophys. Research **64**, 1721 (1959).

H. C. Van de Hulst (1947), Astrophys. J. 105, 471 (1947).

E. Vilcsek and H. Wänke (1961) (to be published); see also Z. Naturforsch. **16a**, 379 (1961).

E. Vilcsek and H. Wänke (1960), Z. Naturforsch. **15a**, 1004 (1960).

A. P. Vinogradov, I. K. Zadorozhnii, and K. G. Knorre (1960), Meteoritika **18**, 92 (1960).

R. Vogel (1961) Chem. Erde **21**, 24 (1961).

H. Voshage and H. Hintenberger (1959), Z. Naturforsch. **14a**, 194 and 828 (1959).

H. Voshage and H. Hintenberger (1960), Nature **185**, 88 (1960).

H. Voshage and H. Hintenberger (1961), Z. Naturforsch. **16a**, 1042 (1961).

H. Wänke (1960a), Nature **188**, 1101 (1960).

H. Wänke (1960b) Lecture at Varenna Summer School on Nuclear Geology, September, 1960.

H. Wänke and H. König (1959), Z. Naturforsch. **14a**, 860 (1959).

H. Wänke and E. Vilcsek (1959), Z. Naturforsch. **15a**, 929 (1959).

G. J. Wasserburg, W. A. Fowler, and F. Hoyle (1960), Phys. Rev. Letters **4**, 112 (1960).

G. J. Wasserburg and R. J. Hayden (1955a), Phys. Rev. **97**, 86 (1955).

G. J. Wasserburg and R. J. Hayden (1955b), Nature **176**, 130 (1955).

G. J. Wasserburg (1954), in *Nuclear Geology*, edited by H. Faul (John Wiley & Sons, Inc., New York, 1954).

G. J. Wasserburg, R. J. Hayden, and K. J. Jensen (1956), Geochim. et Cosmochim. Acta **10**, 153 (1956).

R. K. Webster, J. W. Morgan, and A. A. Smales (1958), Geochim. et Cosmochim. Acta **15**, 150 (1958).

F. L. Whipple and E. L. Fireman (1959), Nature **183**, 1315 (1958).

J. Zähringer and W. Gentner (1960), Z. Naturforsch. **15a**, 600 (1960).

J. Zähringer and W. Gentner (1961), Z. Naturforsch. **16a**, 239 (1961).