# Neutron Exposure Ages of Meteorites

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(Z. Naturforschg. 21 a, 1138—1146 [1966]; received 21 March 1966)

*Dedicated to Professor* Dr. W. GENTNER's *60. anniversary*

$Co^{60}$ and $Ar^{39}$ were measured in a number of recently fallen meteorites. From these data the amount of shielding surrounding the samples in space was estimated. In the carbonaceous chondrite, Murray, the $Co^{60}$ activity was very large compared to the $Co^{60}$ activities in the other chondrites even though the $Ar^{39}$ activities were almost identical. In the iron meteorites there was an inverse relationship between the $Co^{60}$ and $Ar^{39}$ activities; shielding material increased the $Co^{60}$ activity and decreased the $Ar^{39}$ activity.

In the cases of Abee and Bruderheim, neutron exposure ages were obtained from the $Kr^{80}/Co^{60}$, $Kr^{82}/Co^{60}$, and $Xe^{128}/Co^{60}$ ratios because the $Co^{60}$ was measured in large samples from the interior of the same fragment in which krypton, xenon, bromine, and iodine had previously been measured. The neutron exposure ages were approximately an order of magnitude larger than the spallation exposure ages. The order-of-magnitude differences in the exposure ages could be attributed to space erosion or to an ancient neutron irradiation.

The subject of cosmic-ray-produced radioactive and stable isotopes in meteorites has been studied for a decade and is discussed in several excellent review articles [1–3]. Most of these isotopes are produced by spallation, the fragmentation of nuclei caused by the high-energy protons interacting with the material. The relative amounts of the spallation isotopes are to a good approximation in accord with expectations for a constant flux of high-energy protons. The ratio of a spallation-produced stable isotope to a spallation-produced radioactive isotope gives the time of exposure to high-energy protons; this time is called the exposure age. The exposure ages are usually of the order of $10^7$ years in stony meteorites and of the order of $10^8$ years in iron meteorites. These exposure ages can be interpreted both by the breakoff of many meters of material in a single collision and by space erosion.

Radioactive and stable isotopes can also be produced in meteorites by the capture of secondary neutrons that arise from the action of cosmic rays on the material. FIREMAN and SCHWARZER [4] first studied this problem and showed that the amount of $He^3$ produced by neutron capture in $Li^6$ is small compared to spallation $He^3$. GOEL [5] found $Cl^{36}$ in one sample of Canon Diablo produced by neutron capture in $Cl^{35}$. VAN DILLA et al.[6] found that the $Co^{60}$ activity in some meteorites was mainly due to neutron capture in cobalt. Both the spallation-produced and the neutron-produced radioactivities are indicators of the amount of shielding surrounding the sample in space. EBERHARDT et al.[7] calculated the $Co^{60}$ production in spherical bodies of chondrite composition. We measured a spallation-produced isotope, $Ar^{39}$, and a neutron-capture-produced isotope, $Co^{60}$, in adjacent samples of a number of meteorites, and calculated the $Co^{60}$ and $Ar^{39}$ production in iron meteorites to check whether both indicators correspond to the same amount of shielding.

CLARKE and THODE [8, 9] discovered that the $Kr^{80}$ and $Kr^{82}$ anomalies in the Abee meteorite were produced by neutron captures in $Br^{79}$ and $Br^{81}$, and that the $Xe^{128}$ excess was possibly produced by neutron capture in $I^{127}$. MARTI et al.[10] confirmed and established these results and pointed out that the neutrons captured in $Br^{79}$, $Br^{81}$, and $I^{127}$ were resonance neutrons with approximately 100 ev energy.

1 J. R. ARNOLD, Ann. Rev. Nucl. Sci. **11**, 349 [1961].
2 E. ANDERS, Rev. Mod. Phys. **34**, 287 [1962].
3 O. A. SCHAEFFER, Ann. Rev. Phys. Chem. **13**, 151 [1962].
4 E. L. FIREMAN and D. SCHWARZER, Geochim. Cosmochim. Acta **11**, 252 [1957].
5 P. S. GOEL, Cosmogenic $Cl^{36}$ in Iron Meteorites, Carnegie Inst. Tech. Prog. Rept. 1961–1962 [1962].
6 M. A. VAN DILLA, J. R. ARNOLD, and E. C. ANDERSON, Geochim. Cosmochim. Acta **20**, 115 [1960].
7 P. EBERHARDT, J. GEISS, and H. LUTZ, Earth Science and Meteorites, North-Holland Publishing Company, Amsterdam 1963, p. 143.
8 W. B. CLARKE and H. G. THODE, J. Geophys. Res. **69**, 3673 [1964].
9 W. B. CLARKE and H. G. THODE, Isotope Anomalies in Xenon from Meteorites and Xenon from Natural Gases, ed. by H. CRAIG, S. L. MILLER, and G. J. WASSERBURG, Isotope and Cosmic Chemistry, North-Holland Publishing Company, Amsterdam 1964, p. 471.
10 K. MARTI, P. EBERHARDT, and J. GEISS, Z. Naturforschg. **21 a**, 398 [1966].

We measured, in an adjacent sample of Abee, the radioactive isotope $Co^{60}$, which is produced by the capture of resonance neutrons of 132 ev energy. This measurement permits the determination of neutron exposure ages from the ratios of stable to radioactive isotopes produced by the capture of resonance neutrons. We also measured the $Co^{60}$ in Bruderheim material adjacent to that used by MERRIHUE [11] for his krypton and xenon measurements. The neutron exposure age is related to space erosion. If the neutron and spallation exposure ages are identical, there is no space erosion. On the other hand, if the neutron exposure age is larger than the spallation exposure age, the difference provides a measure of the amount of space erosion. The neutron exposure ages also provide information about ancient neutron irradiations and solar flares.

## Measurements

We measured the $Co^{60}$ activity by the gamma-gamma coincidence method with the same equipment previously used for $Al^{26}$ measurements in meteorites and dust [12, 13]. The unit consisted of two pais of NaI(Tl)

← Table 1. $Co^{60}$ and $Ar^{39}$ Activities

| Date of Fall | Meteorites | Wgt (g) | Dates of Counting | $Co^{60}$ (dpm/kg) | $Co^{60}(t_0)$ * (dpm/kg) | $Co^{60}$ (by others) (dpm/kg) | $Ar^{39}$ (dpm/kg) | $Ar^{39}$ (by others) (dpm/kg) |
|---|---|---|---|---|---|---|---|---|
| | *Stones* | | | | | | | |
| June 10, 1952 | Abee | 1124 | October 1964 | 1.7 ± 0.7 | 8.1 ± 3.4 | — | — | 18.6 ± 0.6+ [a] |
| March 4, 1960 | Bruderheim | 1000 | December 1964 | 4.2 ± 0.6 | 7.9 ± 1.1 | 10 ± 1 [b] | 10 ± 1 | — |
| October 13, 1959 | Hamlet | 697 | September 1963 | 3.8 ± 1.5 | 6.5 ± 2.4 | — | 7 ± 1 | 7.6 ± 0.2 [c] |
| April 7, 1959 | Příbram | 144 | September 1965 | < 3.8 | < 9 | — | 6 ± 1 | — |
| September 20, 1950 | Murray | 369 | January 1966 | 18.3 ± 3.3 | 135 ± 24 | — | — | 9.4 ± 0.5 [c] |
| | *Irons* | | | | | | | |
| November 24, 1959 | Aroos | 71 | September 1963 | 20 ± 5 | 33 ± 8 | 17 ± 2 [d] | 16 ± 2 | 16.3 ± 0.9 [e]<br>17.2 ± 0.5 [f] |
| August 14, 1962 | Bogou | 301 | August 1965 and October 1963 | 7.1 ± 1.5, 7.4 ± 1.5 | 9.5 ± 1.5 | <7 ± 4 [g]<br>14 ± 7 [h] | 23 ± 1 | ~ 22 [e]<br>— |
| April 10, 1964 | Muzzafapur | 90 | April 1965 and June 1964 | 9 ± 5, 10 ± 5 | 10 ± 5 | — | 23 ± 1 | — |
| — | Hoba West | 1574 | November 1963 | 2.9 ± 1.0 | — | — | — | — |
| May 10, 1931 | Eaton (copper) | 30 | December 1963 | < 3.3 | < 240 | — | — | — |
| — | Carbo | 1200 | May 1964 | ≦ 1.5 | — | — | < 0.4 | — |
| December 12, 1947 | Sikhote-Alin | 1198 | July 1965 and March 1963 | 30 ± 3, 52 ± 3 | 390 ± 40 | 207 ± 21 [i]<br>386 ± 39 [i]<br>196 ± 4 [j]<br>290 ± 5 [j]<br>225 ± 6 [j]<br>289 ± 30 [j]<br>299 ± 22 [j]<br>320 ± 32 [j] | 5.0 ± 0.4 | 7.07 ± 0.23 [f]<br>7.0 ± 0.3 [k] |

* $t_0$ = time of fall; [a] COBB [14], + metal phase; [b] HONDA et al. [15]; [c] STOENNER et al. [16]; [d] HONDA and ARNOLD [17]; [e] DAVIS et al. [18]; [f] VILCSEK and WÄNKE [19]; [g] ROWE et al. [20]; [h] TOBAILEM and NORDEMANN [21]; [i] ROWE et al. [22]; [j] KAYE [23]; [k] SPRENKEL [24].

11 C. M. MERRIHUE, J. Geophys. Res. **71**, 263 [1966].

12 J. DEFELICE, G. G. FAZIO, and E. L. FIREMAN, Science **142**, 673 [1963].

13 E. L. FIREMAN and C. C. LANGWAY, JR., Geochim. Cosmochim. Acta **29**, 21 [1965].

14 J. C. COBB, Exposure Ages of Some Chondrites from $Ar^{39}$ and $Ar^{38}$ Measurements, Brookhaven National Laboratory Preprint 8915 [1965].

15 M. HONDA, S. UNEMOTA, and J. R. ARNOLD, J. Geophys. Res. **66**, 3541 [1961].

16 R. W. STOENNER, O. A. SCHAEFFER, and R. DAVIS, JR., J. Geophys. Res. **65**, 3025 [1960].

17 M. HONDA and J. R. ARNOLD, Geochim. Cosmochim. Acta **23**, 219 [1961].

18 R. DAVIS, JR., R. W. STOENNER, and O. A. SCHAEFFER, Cosmic-Ray Produced $Ar^{37}$ and $Ar^{39}$ Activities in Recently Fallen Meteorites, Radioactive Dating IAEA, Vienna 1963, p. 355.

19 E. VILCSEK and H. WÄNKE, Cosmic-Ray Exposure Ages, Radioactive Dating IAEA, Vienna 1963, p. 381.

20 M. N. ROWE, E. C. ANDERSON, and M. A. VAN DILLA, J. Geophys. Res. **69**, 521 [1964].

21 J. TOBAILEM and D. NORDEMANN, Geochim. Cosmochim. Acta **29**, 1317 [1965].

22 M. N. ROWE, M. A. VAN DILLA, and E. C. ANDERSON, Geochim. Cosmochim. Acta **27**, 1003 [1963].

23 J. H. KAYE, Cosmogenic X-Ray and $\beta$-Ray Emitters in Iron Meteorites, Thesis, Carnegie Inst. Tech. 1963.

24 E. SPRENKEL, Cosmic Ray Produced Cl-36 Activities in Iron Meteorites, Thesis, Univ. of Rochester 1959.

activated crystals enclosed in an 8-inch-thick zinc shield. The crystals in one pair were 6 inches in diameter and 4 inches thick with a 10% resolution, and in the other pair 3 inches in diameter and 3 inches thick with an 8% resolution. Most $Co^{60}$ measurements were made with the 6- by 4-inch crystals because of their higher efficiency for the 1.17- and 1.33-Mev gamma rays. In normal operation the pulse from one crystal fed a linear amplifier with the discriminator set at $(1.25 \pm 0.10)$ Mev. The output of the discriminator fed the trigger input of a TMC analyzer. After an appropriate delay of 2 $\mu$sec the pulse from the other crystal was fed into the signal input of the analyzer. The analyzer covered the range from (0.2 to 2.5) Mev. The sample and background were counted for times between 1 and $2 \times 10^4$ min. The background was determined with a blank specimen between the crystals in the same geometry. To determine the background for the stony meteorites we used a blank specimen consisting of a mixture of NaCl and Fe powder in a mold made from the sample. For the iron meteorites we used a set of iron sheets in the shape of the sample. We prepared calibration samples by uniformly distributing a known amount of $Co^{60}$ activity in the NaCl-Fe powder for the stony meteorites, and on blotting paper placed next to the iron sheets for the iron meteorites.

Fig. 1 shows the $Co^{60}$ activities for Sikhote-Alin and Bogou. The $Co^{60}$ activity was measured in 1963 and in 1965. The decay in the $Co^{60}$ activity was consistent with the half-life of $Co^{60}$.

## Relation of $Co^{60}$ and $Ar^{39}$ to the Chemical Composition and Size of Meteorites

We found the $Co^{60}$ activities in Murray, Abee, Bruderheim, Hamlet and Přı́bram to be $135 \pm 24$, $8.1 \pm 3.4$, $7.9 \pm 1.1$, $6.5 \pm 2.4$, and less than 9 dpm/kg, respectively (see Table 1). The large $Co^{60}$ activity in Murray is probably due to the amount of hydrogen in the material, hydrogen being a very efficient neutron moderator. Wiik[25] found 12.4% of $H_2O$ in Murray. We measured the $Ar^{39}$ activities (by the method described by Fireman and DeFelice[26]) in adjacent samples of Bruderheim, Hamlet, and Přı́bram and obtained the values $10 \pm 1$, $7 \pm 1$, and $6 \pm 1$ dpm/kg, respectively. The $Ar^{39}$ in Murray was found to be $9.4 \pm 0.5$ dpm/kg[16], and in Abee to be $18.6 \pm 0.6$ dpm/kg in the metal phase. Except for the high $Co^{60}$ activity in Murray there is little variation in the $Co^{60}$ activities in the stony meteorites. The $Co^{60}$ activities are less than those calculated by Eberhardt et al.[27] at the center of a sphere of 40-cm radius. The $Ar^{39}$ activities in the whole rock samples vary by less than a factor of 2. These $Ar^{39}$ activities indicate that the amounts of shielding that surrounded the samples in space differed by less than

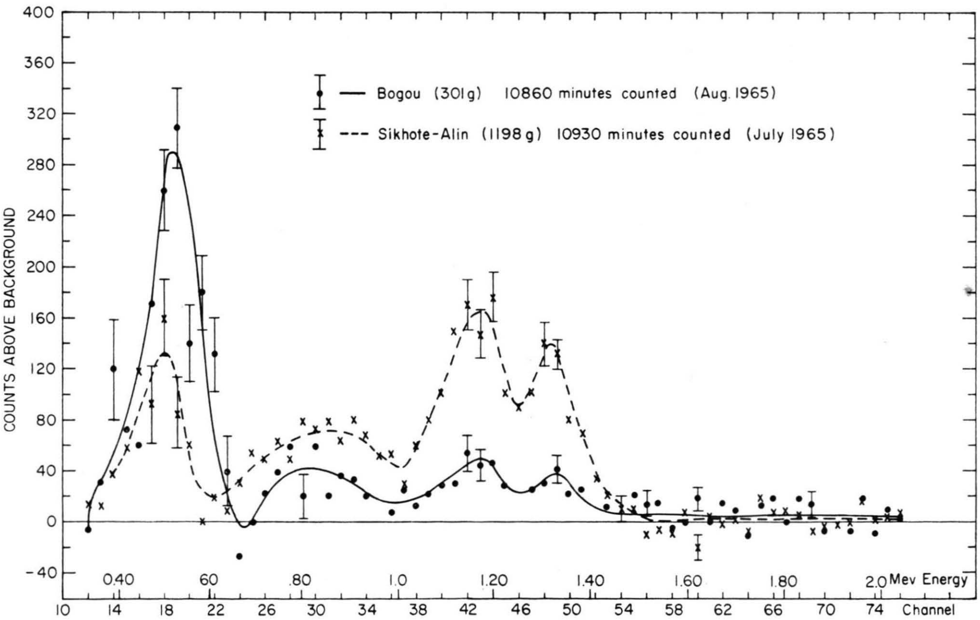


Fig. 1. Gamma-ray spectrum in coincidence with $1.25 \pm 0.10$ Mev gamma rays for Bogou and Sikhote-Alin.

[25] H. B. Wiik, Geochim. Cosmochim. Acta **9**, 279 [1956].

[26] E. L. Fireman and J. DeFelice, Geochim. Cosmochim. Acta **18**, 183 [1960].

[27] See ref. [7] (p. 1138).

40 cm. Evidently for stony meteorites the $Co^{60}$ and $Ar^{39}$ are affected more by differences in chemical composition than by differences in size.

The $Co^{60}$ activity in iron meteorites, on the other hand, is simply related to the $Ar^{39}$ activity. The $Co^{60}$ and $Ar^{39}$ activities in neighboring samples of Aroos, Bogou, Muzzafapur, and Sikhote-Alin are ($33 \pm 8$, $16 \pm 2$), ($9.5 \pm 1.5$, $23 \pm 1$), ($10 \pm 5$, $23 \pm 1$), and ($390 \pm 40$, $5.0 \pm 0.4$) dpm/kg, respectively, at the time of fall. The simple inverse relation between $Ar^{39}$ and $Co^{60}$ indicates that as the proton flux which produces $Ar^{39}$ decreases, the neutron flux which produces the $Co^{60}$ increases. Bogou and Muzzafapur, with the highest $Ar^{39}$ activities, ($23 \pm 1$) dpm/kg, had the lowest $Co^{60}$ activities, ($9.5 \pm 1.5$) and ($10 \pm 5$) dpm/kg. Sikhote-Alin, with the lowest $Ar^{39}$ activity, ($5.0 \pm 0.4$) dpm/kg, had the highest $Co^{60}$, ($390 \pm 40$) dpm/kg. Aroos, with the intermediate $Ar^{39}$ activity, ($16 \pm 2$) dpm/kg, had the intermediate $Co^{60}$ activity, ($33 \pm 8$) dpm/kg. Table 1 gives our values together with previous measurements on the same meteorites.

The calculations for spheres given in Appendix A and illustrated in Fig. A 1, show that the amount of shielding necessary for the $Ar^{39}$ activity is somewhat larger than that necessary for the $Co^{60}$. In Bogou and Muzzafapur the $Ar^{39}$ corresponds to between 20 and 30 cm of shielding, and the $Co^{60}$ activity to less than 20 cm of shielding. In Aroos the $Ar^{39}$ corresponds to between 35 and 43 cm of shielding and the $Co^{60}$ to between 26 and 32 cm of shielding. In Sikhote-Alin the $Ar^{39}$ corresponds to 70 cm of shielding and the $Co^{60}$ to 55 cm of shielding. The differences may be due to deviations from sphericity.

Table 1 also gives the $Co^{60}$ results in two non-recently fallen iron meteorites, Hoba West and Carbo, and in a suspected copper meteorite, Eaton. There was a small amount of $Co^{60}$ in Hoba West, ($2.9 \pm 1.0$) dpm/kg. Since a small amount of $Co^{60}$ can arise from $Fe^{60}$, which has a $10^5$ year half-life [28], the activity in Hoba West may be caused by its $Fe^{60}$ content. The Eaton copper sample was said to have fallen on May 30, 1931 [29], which was approximately six half-lives of $Co^{60}$ before our measurement. No $Co^{60}$ was observed in Eaton; our limit corresponds to less than 240 dpm/kg at the time of fall. Since the cosmic-ray-production cross section of $Co^{60}$ in copper is large ($\sim 100$ mb), the lack of $Co^{60}$ in the sample sets an upper limit to the cosmic-ray bombardment flux of 3 $cm^{-2}$ $sec^{-1}$, which is slightly less than the 5 $cm^{-2}$ $sec^{-1}$ value expected for a small copper body in space.

## The Neutron Exposure Ages of Abee and Bruderheim

The $Co^{60}$ activity that we measured in Abee can be combined with the $Kr^{80}$, $Kr^{82}$, and $X^{128}$ excesses obtained by CLARKE and THODE [8, 9] and MARTI et al. [10] and the bromine and iodine abundances obtained by MERRIHUE [11] to derive neutron exposure ages, because the $Co^{60}$ was measured in an interior slice of large area. The slice was rectangular in shape, with sides $23 \times 10$ cm, and with a weight of 1124 g. Since only one fragment of Abee was recovered, our $Co^{60}$ value of ($8.1 \pm 3.4$) dpm/kg should be representative of the region where the krypton and xenon excesses were measured. In Bruderheim we measured the $Co^{60}$ in a 210-g magnetic fraction separated from a 1-kg interior sample of B-74 (Folinsbee's fragment classification). This fraction was taken from the same sample used by MERRIHUE [11] for his measurements. Since many fragments of Bruderheim exist, the $Co^{60}$ activity may differ in other fragments. Our $Co^{60}$ value of ($7.9 \pm 1.1$) dpm/kg is strictly applicable only to MERRIHUE's sample. However, if CLARKE and THODE [8, 9] and MARTI et al. [10] used samples from fragment B-74, our result would also apply to their samples.

Although $Ar^{36}$ has been discussed as a possible neutron-capture-produced isotope from $Cl^{35}$, we have not compared the $Co^{60}$ activities with $Ar^{36}$ excesses for the following reasons: (1) the energy dependence of the neutron-capture cross section in $Cl^{35}$, which lacks resonances, is different from that in cobalt; (2) there are only two argon isotopes, $Ar^{36}$ and $Ar^{38}$, that can be used to separate large spallation and primordial contributions from neutron-capture-produced $Ar^{36}$; (3) argon diffusion loss from the chlorine-containing minerals could be serious, and (4) the $Ar^{36}$ excess expected from neutron capture in chlorine is very small.

The production rate of $Kr^{80}$, $Kr^{82}$, and $Xe^{128}$ can be obtained from the $Co^{60}$ activity because these isotopes all have large neutron-capture resonances between 20 and 300 ev. Furthermore, the cobalt

[28] P. S. GOEL and M. HONDA, J. Geophys. Res. **70**, 747 [1965].

[29] H. H. NININGER, Pop. Astron. (Contr. Soc. Res. Meteorites) **51**, 273 [1943].

content of meteorites is known. The neutron flux and energy spectrum in a meteoroid depend in a sensitive way on the size of the body and the location of the sample [7]. However, the $Kr^{80}/Co^{60}$, $Kr^{82}/Co^{60}$, and $Xe^{128}/Co^{60}$ ratios should be approximately independent of the size of the meteoroid and the sample location (see Appendix B).

The neutron exposure ages, $\tau_N(80)$, $\tau_N(82)$, and $\tau_N(128)$, are given by the following expressions:

$$\tau_N(80) = \frac{Kr^{80}\text{ excess}}{r_{80}\, Co^{60}} \quad \text{where } r_{80} = (1.30)\frac{\#\, Br^{79}}{\#\, Co^{59}},$$

$$\tau_N(82) = \frac{Kr^{82}\text{ excess}}{r_{82}\, Co^{60}} \quad \text{where } r_{82} = (0.55)\frac{\#\, Br^{81}}{\#\, Co^{59}},$$

$$\tau_N(128) = \frac{Xe^{128}\text{ excess}}{r_{128}\, Co^{60}} \quad \text{where } r_{128} = (1.69)\frac{\#\, I^{127}}{\#\, Co^{59}}.$$

The symbol # denotes the number of atoms/cm$^3$. The production ratios of $Kr^{80}$, $Kr^{82}$, and $Xe^{128}$ to $Co^{60}$, denoted by $r_{80}$, $r_{82}$, $r_{128}$ are calculated in Appendix B. The cobalt content is 700 ppm.

The $Kr^{80}$ and $Kr^{82}$ excesses based on $Kr^{86}$ normalization and the bromine contents taken from Merrihue's [11] measurements are given in Table 2. The neutron exposure ages obtained from the $Kr^{80}$, $Kr^{82}$, and $Xe^{128}$ excesses and the $Co^{60}$ activity are also given in Table 2. For Abee the $Kr^{80}$ excess of $(4.0 \pm 0.2) \times 10^8$ atoms/g combined with the $Co^{60}$ activity of $8.1 \pm 3.4$ dpm/kg gives a $Kr^{80}/Co^{60}$ age of (37 ) $\times 10^6$ years; the $Kr^{80}$ excess of $(5.7 \pm 0.2) \times 10^8$ atoms/g combined with the $Co^{60}$ activity gives a $Kr^{80}/Co^{60}$ age of (53 ) $\times 10^6$ years. The minimum and maximum $Kr^{80}/Co^{60}$ ages for Abee are 25 and $96 \times 10^6$ years. These neutron exposure ages are between 3.8 and 14.5 times greater than the spallation exposure age of $6.6 \times 10^6$ years. The $Kr^{82}$ excesses of 1.7 and $2.7 \times 10^8$ atoms/g combine with the $Co^{60}$ to give $Kr^{82}/Co^{60}$ ages of 39 and $62 \times 10^6$ years. The $Kr^{80}/Co^{60}$ and the $Kr^{82}/Co^{60}$ ages in Abee are approximately an order of magnitude larger than the spallation exposure age.

In the Bruderheim material adjacent to the sample in which we measured the $Co^{60}$, the bromine content was very low, 0.02 ppm in the whole rock and 0.072 ppm in a chondrule [11]. It is therefore not surprising that the neutron-capture-produced $Kr^{80}$ is small for the whole-rock samples. There is an indication for neutron-capture-produced krypton in the chondrules. If the $Kr^{80}$ and $Kr^{82}$ excesses in chondrule BC-1, which had a relatively low $Kr^{83}$ excess, are interpreted as neutron capture in bromine, then $Kr^{80}/Co^{60}$ and $Kr^{82}/Co^{60}$ neutron exposure ages of approximately $400 \times 10^6$ years result for Bruderheim; these neutron exposure ages are more than an order of magnitude greater than the spallation exposure age of $26 \times 10^6$ yeas. Because of the uncertainties in the krypton in chondrule BC-1 and its small bromine content, the $Kr^{80}/Co^{60}$ ages for Bruderheim are very uncertain.

It is more difficult to estimate the neutron-capture-produced $Xe^{128}$. The $Xe^{128}$ excess in a meteorite depends critically on which xenon isotope is chosen for normalization, the isotopic composition of primordial xenon, and the amount of spallation xenon. If the xenon is normalized at $Xe^{130}$, if the $Xe^{128}/Xe^{130}$ ratio in primordial xenon is the same as in the atmosphere, and if the spallation $Xe^{128}$ equals the $Xe^{126}$ content, then we obtain the values of the $Xe^{128}$ and $Xe^{128}/Co^{60}$ ages given in Table 2. The $Xe^{128}$ excess in Abee is more than an order of

| Meteorite | Excess in $10^8$ atoms/g | | | | | Exposure Ages in units of $10^6$ years | | | |
|---|---|---|---|---|---|---|---|---|---|
| | $Kr^{80}$ | $Kr^{82}$ | $Xe^{128}$ | Br (ppm) | I (ppm) | $Kr^{80}/Co^{60}$ | $Kr^{82}/Co^{60}$ | $Xe^{128}/Co^{60}$ | $(Ar^{38}/Ar^{39})$ metal |
| Abee | $4.0 \pm 0.2$ [a] | $1.7 \pm 0.7$ [a] | 3.9 [b] | 3.6 [c] | 0.22 [c,d] | 37 | 39 | ~ 390 | 6.6 [e] |
| Abee | $5.7 \pm 0.2$ [f] | $2.7 \pm 0.7$ [f] | 0.91 [f] | 3.6 [c] | 0.22 [c,d] | 53 | 62 | ~ 92 | 6.6 [e] |
| Bruderheim Chondrule (BC—1) | $0.68 \pm 0.27$ [c] | $0.37 \pm 0.13$ [c] | — | 0.072 [c] | 0.0023 [c] | ~400 | ~400 | — | 26 [e] |
| Bruderheim | $0.21 \pm 0.01$ [a] | $0.32 \pm 0.01$ [a] | 2.3 [b] | 0.02 [c] | 0.012 [c,d] | — | — | ~1200 | 26 [e] |
| Bruderheim | 0.95 [f] | 1.24 [f] | — | 0.02 [c] | 0.012 [c,d] | — | — | — | — |

Table 2. Neutron Exposure Ages. [a] Clarke and Thode [8]; [b] Clarke and Thode [9]; [c] Merrihue [11]; [d] Goles and Anders [30]; [e] Cobb [14]; [f] Marti et al. [10].

[29] K. R. Dawson, J. A. Maxwell, and D. E. Parsons, Geochim. Cosmochim. Acta **21**, 127 [1960].

[30] G. G. Goles and E. Anders, Geochim. Cosmochim. Acta **26**, 723 [1962].

magnitude larger than would be obtained from the $Co^{60}$ activity and the spallation exposure age. The $Xe^{128}/Co^{60}$ age for Abee ranges from 92 to $390 \times 10^6$ years. On the basis of xenon normalization at $Xe^{132}$ and different assumptions concerning primordial and spallation xenon, MARTI et al. [31] obtained a $Xe^{128}$ excess of $0.68 \times 10^8$ atoms/g, which gives a $Xe^{128}/Co^{60}$ age of $69 \times 10^6$ years for Abee.

In the carbonaceous chondrite, Murray, the amounts of neutron-produced $Kr^{80}$, $Kr^{82}$, and $Xe^{128}$ are small compared to the amounts from other origins.

The $Kr^{80}/Co^{60}$, $Kr^{82}/Co^{60}$, and $Xe^{128}/Co^{60}$ ages in Abee and also in Bruderheim require a neutron dosage an order of magnitude larger than that given by the product of the recent neutron flux and the spallation exposure age. The neutron dosage could be due to an ancient neutron irradiation during the early history of the solar system, or it could be due to space erosion with a collision more ancient than that given by the spallation exposure age. It is easy to make the neutron exposure age an order of magnitude larger than the spallation exposure age by space erosion. The relation between the spallation exposure age $E$, the erosion rate $\varepsilon$, the cosmic-ray absorption length $\mu$ (which is taken to be 50 cm for a chondrite), and the collision time $T$ is [32]

$$E = \frac{\mu}{\varepsilon} [1 - e^{-\varepsilon T/\mu}].$$

If for Abee $\varepsilon = 4 \times 10^{-6}$ cm/yr and $T = 10^7$ years, then the spallation exposure age $E$ would be $6.9 \times 10^6$ years and the neutron exposure age would be approximately $40 \times 10^6$ years. If space erosion were the dominating process, then $\varepsilon$ would be $7.6 \times 10^{-6}$ cm/yr and the neutron exposure age would be approximately $130 \times 10^6$ years.

If the neutron exposure age and the spallation exposure age are different, as indicated by the low $Co^{60}$ activities in Abee and Bruderheim, then the collision times of meteorites are less frequent than given by the spallation exposure ages. The equivalence of collision times and spallation exposure ages implies, according to a recent calculation [33], that collisions are so frequent for asteroidal meteoroids that the brightness of the night sky is increased. The concentration of dust required for space erosion [26] is consistent with the intensity of the zodiacal light.

[31] See ref. [10] (p. 1138).
[32] F. L. WHIPPLE and E. L. FIREMAN, Nature (London) **183**, 1315 [1959].

*Acknowledgments.* I wish to thank Mr. J. D'AMICO for his assistance with many of the measurements and for maintaining the electronic equipment. I am grateful to Mr. J. DEFELICE for his help and to Mr. J. KORNBLUM for his assistance with the computations. The meteorite samples used in this work were loaned or donated by Mr. ROY S. CLARKE, JR. and Dr. E. HENDERSON of the U.S. National Museum, Dr. Z. CEPLECHA of the Ondřejov Observatory, Prof. R. E. FOLINSBEE of the University of Alberta, and Prof. E. L. KRINOV of the USSR Committee on meteorites.

I also wish to acknowledge helpful discussions with Dr. HENRI MITLER.

## Appendix A

### *Calculation of $Co^{60}$ in Iron Meteorites from the $Ar^{39}$ Content*

The relative amounts of cosmic-ray-produced $Ar^{39}$ in iron spheres of different sizes can be obtained from the measured production cross sections and depth variation, and from the cosmic-ray energy spectrum. If the $Ar^{39}$ activity is normalized to a maximum of 27 dpm/kg, we obtain the dashed curve in Fig. A 1 and the solid curves given in Fig. A 2 for the $Ar^{39}$ activity in iron spheres of different sizes. The $Ar^{39}$ activity is not only a measure of depth of the sample in the meteoroid and the size of the meteoroid but also of the neutron production. Since 1 $Ar^{39}$ is produced in 100 iron spallations and approximately 4 neutrons are produced in each spallation, approximately 400 neutrons are produced for each $Ar^{39}$. The neutrons are usually divided into knock-on energetic neutrons with energies greater than 10 Mev and evaporation neutrons with energies of a few Mev. There is approximately one knock-on and three evaporation neutrons per spallation. The knock-on neutrons with very great energies ($>$ 300 Mev) are indistinguishable from protons of the same energy. The knock-on neutrons with energies in the 10 to 30 Mev range have a large probability for producing $Co^{60}$ by the (n, p) reaction on $Ni^{60}$. Both evaporation and knock-on neutrons can produce $Co^{60}$ by being captured in $Co^{59}$, particularly after their energies are reduced. Before we calculate the neutron-capture probability, let us calculate the probability of $Co^{60}$ production from $Ni^{60}$.

The (n, p) cross section is large for neutrons with energies between 10 and 30 Mev (approximately 200 mb) and small for neutrons of other energies. The 10- to 30-Mev neutron flux at the center of iron spheres is estimated in the following way. The mean free path for inelastic collisions of neutrons of energy greater than 10 Mev in iron is approximately 10 cm. The flux of knock-on neutrons at the center of a sphere that

[33] J. R. ARNOLD, Astrophys. J. **141**, 1548 [1965].

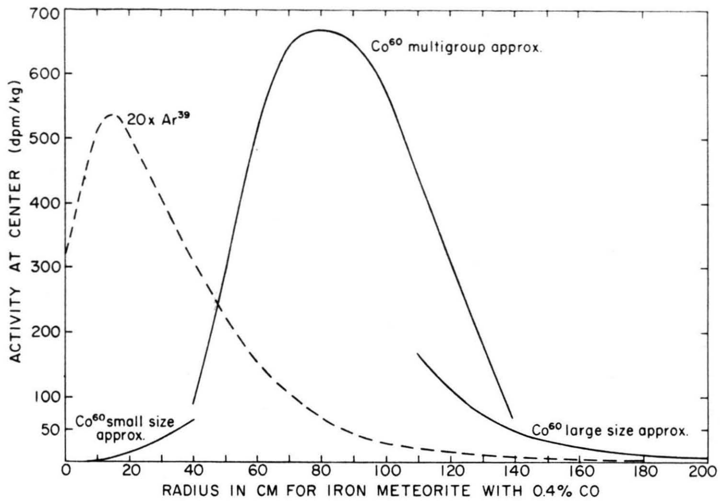


Fig. A1. $Ar^{39}$ and $Co^{60}$ activities at the centers of iron spheres of different radii. (On the abscissa read Co instead of CO.)

have not suffered any inelastic collisions is

$$p \int_0^R e^{-r/10}\, dr = 10\, p\, (1 - e^{-R/10}),$$

where $p$ is the average production rate of knock-on neutrons, $10^2\, \overline{Ar^{39}}$, and $R$ is the radius of the sphere. The 10- to 30-Mev neutron flux is the sum of contributions from knock-on neutrons that have made zero, one, two, three, etc., inelastic collisions. This flux can be written approximately as

$$10 \left[ p_0 (1 - e^{-R/10}) + \frac{p_1}{2} (1 - e^{-R/10})^2 + \frac{p_2}{3} (1 - e^{-R/10})^3 + \frac{p_3}{4} (1 - e^{-R/10})^4 \right],$$

where $p_0$, $p_1$, $p_2$, and $p_3$ are the average production rates of neutrons that have the proper energy, 10 to 30 Mev, after the collisions [34]. If the energy spectrum of the knock-on neutrons is $C/(E+50)$, where $C$ is a constant and $E$ is the energy of the neutron in Mev [36], and if the neutrons lose two-thirds of their energy in an inelastic collision with iron, then the 10- to 30-Mev neutron flux, $F(10-30)$, at the center of the sphere, is

$$F(10-30) = 10^3\, \overline{Ar^{39}} [0.25 (1 - e^{-R/10}) + 0.07 (1 - e^{-R/10})^2 + 0.12 (1 - e^{-R/10})^3 + 0.06 (1 - e^{-R/10})^4].$$

If $F(10-30)$ is in units of $cm^{-2}\, min^{-1}$, then $\overline{Ar^{39}}$ is in units of $cm^{-3}\, min^{-1}$. The $Co^{60}$ production rate is

$$Co^{60}\, (dpm/kg) = F(10-30)\, (2 \times 10^{-25}\, cm^2)\, (\#\, Ni^{60}/kg).$$

For an iron meteorite with 7% nickel, the $Co^{60}$ production rate is $0.15\, \overline{Ar^{39}}$ for $R > 10$ cm. The process accounts for one-third of the $Co^{60}$ observed in Muzzafapur and Bogou, and one-tenth of the $Co^{60}$ in Aroos; however, it could not account for the $Co^{60}$ activity in the stony meteorites because of their lower nickel contents. Let us now consider neutron capture in cobalt.

For neutrons above 2 Mev the inelastic scattering cross section is large and the neutrons are reduced to 2 Mev energy in a few collisions; furthermore, neutron-capture cross sections are small and hence very few captures occur. At approximately 2 Mev the inelastic scattering cross section in iron and nickel becomes small and the elastic scattering cross section becomes large. The values that we use for the elastic scattering cross sections are the total cross sections given by Hughes and Schwartz [37] for iron. The number of elastic collisions, $\nu$, required to change the energy from $E_0$ to $E_1$ is

$$\frac{A + \frac{2}{3}}{2} \ln \frac{E_0}{E_1},$$

where $A$ is the average atomic weight 57, so that 66 elastic collisions are necessary to reduce the energy of a neutron by an order of magnitude, and nearly 500 collisions are necessary to thermalize the neutrons. The average travel distance of a neutron, whose energy is reduced from $2 \times 10^6$ ev to 1 ev, is approximately 40 cm.

[34] The number of neutrons that reach the center after $n$ inelastic collisions is approximately the same as the number of neutrons from an equivalent source at the center that escape from the sphere after $n$ inelastic collisions [35]. The number of neutrons that escape after $n$ inelastic collisions is approximately

$$p_R \int_0^R e^{-r/l} (1 - e^{-r/l})^n\, dr,$$

where $p_R$ is the average neutron production rate in the sphere and $l$ is the mean free path.

[35] H. A. Bethe, The Reciprocity Theorem in Neutron Scattering, Los Angeles Report 1428 [1952].

[36] B. Rossi, High-Energy Particles, Prentice-Hall Inc., Englewood Cliffs, New Jersey 1961, p. 488.

[37] D. J. Hughes and R. B. Schwartz, Neutron Cross-Sections, Brookhaven National Laboratory Report 325 [1958].

In small spheres, $R < 40$ cm, the neutron flux escaping from the surface is $(400\,\overline{Ar^{39}})\ R/3$, where $\overline{Ar^{39}}$ is the average $Ar^{39}$ production. The neutron flux in the interior of the sphere is approximately $(\bar{\nu}+1)$ $(400\,\overline{Ar^{39}})\ R/3$, where $\bar{\nu}=R^2/l^2$ is the average number of elastic collisions and $l$ is the mean free path, 3.7 cm. The $Co^{60}$ production rate for a small iron meteorite with 0.4% cobalt and with 20 mb for the capture cross section in cobalt is

$$Co^{60}\,(\text{dpm/kg}) = 0.84\times10^{-3}\,R\left(\frac{R^2}{3.7^2}+1\right)\overline{Ar^{39}}\,(\text{dpm/kg}),$$

where $R$ is the radius in cm. For a 30-cm radius the $Co^{60}$ production rate is $1.67\,\overline{Ar^{39}}$ or 35 dpm/kg.

Near the center of large spheres, $R > 100$ cm, an entirely different approximation can be used. Neutrons are captured approximately 40 cm from their point of production and the neutron production rate is practically constant (see Fig. A 2). In an infinite body with a constant neutron production rate, the differential neutron flux, $\Phi(E)$, is

$$\Phi(E) = \frac{C\exp\left\{-\frac{1}{\xi}\int_E^{E_0}\frac{\sigma_a}{\sigma}\frac{dE}{E}\right\}}{E} = \frac{C\,p(E)}{E},$$

where $C$ is a constant, $\xi$ is $2/(A+2/3)$, and $\sigma_a$ and $\sigma$ are the absorption and the total cross sections [38]. Near the center of a large iron meteorite with 0.4% cobalt the ratio of neutron capture in cobalt to the total neutron capture is

$$\frac{Co^{60}}{400\,\overline{Ar^{39}}} = \frac{0.004\int_{E_t}^{E_0}\sigma_a(\text{Co})\,p(E)\,\frac{dE}{E}}{\int_{E_t}^{E_0}\sigma_a\,p(E)\,\frac{dE}{E}} = 0.274.$$

The $Co^{60}$ production rate is $110\,\overline{Ar^{39}}$, where $\overline{Ar^{39}}$ is the $Ar^{39}$ production rate averaged over a radius of 40 cm. The large sphere and the small sphere approximations are plotted in Fig. A 1.

For iron meteorites of intermediate sizes $40 \leq R \leq 120$ cm, the multigroup method described by EBERHARDT et al. [7] for stony meteorites was used for the irons. The source function, $S(r)$, for the neutron production was taken to be $400\,Ar^{39}$, which equals $400\,(\sinh\lambda r/\lambda r)$ $(\lambda R/\sinh\lambda R)$, where $\lambda R = 1.48$, 2.02, 2.56, 3.17, 3.80, 5.07, and 5.75 for $R = 40$, 50, 60, 70, 80, 100, and 120 cm, respectively. Fig. A 2 gives these source functions as dotted curves for $R = 40$, 50, 60, 70, and 80 cm. The source functions agree with 400 times the $Ar^{39}$ production except near the surface. We used 52 b for the neutron-capture resonance integral at 132 ev [39, 40, 41]. The resonance contributes 79% of the $Co^{60}$ production for $R = 40$ cm, and 85% for $R = 60$ cm and greater. Fig. A 1 gives the calculated $Co^{60}$ activities together with the $Ar^{39}$ activities at the center of the spheres.

## Appendix B

*Production Ratios of $Kr^{80}$, $Kr^{82}$, and $Xe^{128}$ to $Co^{60}$ by Neutron Capture*

The neutron-capture cross section in cobalt has been well measured. Its thermal-capture cross section is 36 b [42] and its neutron-capture resonance integral above the cadmium cutoff, 0.5 ev, is $77 \pm 4$ b [39–41]. The thermal-capture cross sections in $Br^{79}$, $Br^{81}$, and iodine are 11.4, 3.1, and 6.2 b, respectively [41]. The measured neutron-capture resonance integral above the cadmium cutoff is 147 b for $Br^{79}$ and 150 b for iodine [43]. The $Br^{79}$ value is 50% higher than the value of 100 b obtained from the recent resonance parameter [44]; the

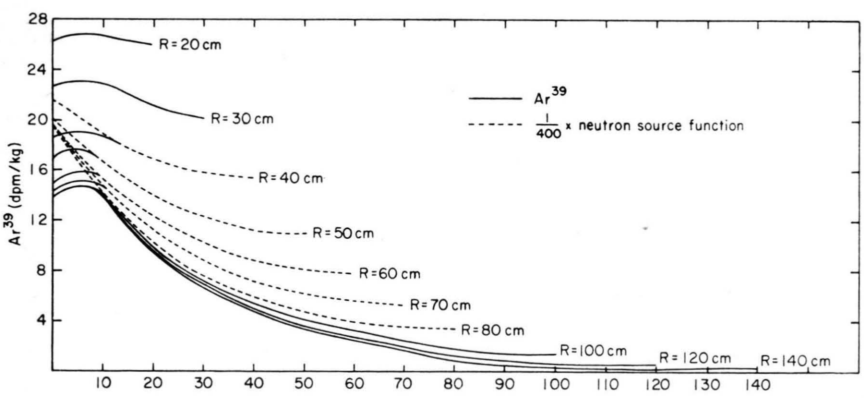


Fig. A 2. $Ar^{39}$ activities and neutron source functions as a function of depth in iron spheres of radii $R$, for $R$ from 20 to 140 cm.

[38] S. GLASSTONE and M. C. EDLUND, The Elements of Nuclear Reactor Theory, D. Van Nostrand Inc., New York 1952.

[39] F. FEINER and L. T. ESCH, Measurements of Resonance Activation Integrals, KAPL 2, 000-12, Dec. [1960].

[40] F. J. JOHNSTON, J. HALPERIN, and R. W. STOUGHTON, J. Nucl. Energy **A 11**, 95 [1960].

[41] R. DAHLBERG, K. JARLOW, and E. JOHANSSON, J. Nucl. Energy **A 14**, 53 [1961].

[42] S. A. COX, Phys. Rev. **133**, B 378 [1964].

[43] R. L. MACKLIN and H. S. POMERANCE, Physics and Mathematics **1**, Pergamon Press, New York 1956, p. 179.

[44] K. H. SAN, L. B. PIKELNER, E. I. SHAROPOV, and K. SIRAZHET, Radiation Width of Nuclei in the Region of Mass Numbers 60–100, Int. Conf. Study of Nuclear Structure E 2214, Dubna, Yugoslavia [1965].

iodine value does not differ significantly from the value of 130 b obtained from the recent resonance parameters of GARG et al.[45]. The $Br^{82}$ neutron-capture integral above the cadmium cutoff is 42 b on the basis of the resonance parameters. The strongest $Br^{81}$ resonances are at 102 ev and 136 ev, the strong cobalt resonance at 132 ev.

The values of the neutron-capture integral above the cadmium cutoff that we shall use are: $77 \pm 4$ b for cobalt, $100 \pm 20$ b for $Br^{79}$, $42 \pm 3$ for $Br^{81}$, and $130 \pm 20$ for iodine. The production ratios of $Kr^{80}$, $Kr^{82}$, and $Xe^{128}$ to $Co^{60}$ by neutron capture are

$$r_{80} = \frac{P_{res}(Br^{79}) + P_{1/v}(Br^{79})}{P_{res}(Co) + P_{1/v}(Co)} \cong \frac{\# \, Br^{79}(100 \pm 20)}{\# \, Co^{59}(77 \pm 4)},$$

$$r_{82} = \frac{P_{res}(Br^{81}) + P_{1/v}(Br^{81})}{P_{res}(Co) + P_{1/v}(Co)} \cong \frac{\# \, Br^{81}(42 \pm 3)}{\# \, Co^{59}(77 \pm 4)},$$

$$r_{128} = \frac{P_{res}(I^{127}) + P_{1/v}(I^{127})}{P_{res}(Co) + P_{1/v}(Co)} \cong \frac{\# \, I^{127}(130 \pm 20)}{\# \, Co^{59}(77 \pm 4)},$$

where $P_{res}$ is the resonance-capture production and $P_{1/v}$ is the sum of the $1/v$ and the thermal-capture productions. The symbol # denotes the number of atoms/$cm^3$. For chondrites, the quantities $P_{res}$ and $P_{1/v}$, and therefore the production ratios, can be evaluated by the method of EBERHARDT et al.[7]. The values for the production ratio $r_{80}$ that we obtain by their procedure are: 1.49 # $Br^{79}$/# $Co^{59}$ for 30 cm of shielding, 1.28 # $Br^{79}$/# $Co^{59}$ for 60 cm of shielding, and 0.94 # $Br^{79}$/# $Co^{59}$ for an infinite of shielding. These values are approximately the same as the ratio of the neutron-capture integrals above the cadmium cutoff, 1.30. The production ratios $r_{82}$ and $r_{128}$ vary less with amounts of shielding than $r_{80}$.

[45] J. B. GARG, W. W. HAVENS, JR., and J. RAINWATER, Phys. Rev. **136**, B 185 [1964].