Radioactive Isotopes in Hoba West and Other Iron Meteorites

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Abstract. The radioactivities Be^{10}, Al^{26}, Cl^{36}, Mn^{53}, Ni^{59}, and Co^{60} and the rare gas isotopes were measured in a sample taken from the surface of the Hoba meteorite. The spallation-produced radioactivities indicate that the sample was at a depth of 35 to 40 cm when the body was in space. The Ni^{59} activity indicates that the terrestrial age of Hoba is less than 80,000 years. Its Cl^{36}-Ar^{36} exposure age is 263±40 million years. The Cl^{36} and rare-gas isotopes were also measured in the Deelfontein meteorite; its Cl^{36}-Ar^{36} exposure age is 400±40 million years. The Cl^{36} and the Ni^{59} were also measured in a sample of Sikhote-Alin. In both Hoba and Sikhote-Alin the Ni^{59}, which is a neutron-produced isotope, is higher than would be expected from the Cl^{36} content. This indicates that solar flares contributed to the neutron-produced isotopes in these iron meteorites.

I. INTRODUCTION

The largest known meteorite fragment is the 60-ton high-nickel ataxite Hoba West, which still lies in place 12 miles west of Grootfontein, South-West Africa, where it was found in 1920 (Prior and Hey 1953, Spencer 1932). The 60-ton weight of the present unweathered mass is estimated from its dimensions, 3 × 3 × 0.9 m (Spencer 1932). A 30 cm thick layer of "iron shale," a weathering product, surrounds the sides and bottom of the meteorite and probably once covered the top. Previous work on this meteorite consisted of chemical analyses of the metal and iron shale (Spencer 1932), a deter-

mination of the He^3 and He^4 (Bauer 1963) and Co^{60} (Fireman 1966) contents, and a determination of the K^{40}-K^{41} cosmic-ray exposure age (Voshage 1967). The cosmic-ray exposure age estimated from the He^3 and He^4 contents (Bauer 1963) is 190 million years; it is difficult to estimate the uncertainty, which is thought to be considerable, in this type of determination. The K^{40}-K^{41} age is 300 million years, with an accuracy of better than ±100 million years (Voshage 1967).

The present study of the radioactivities and rare-gas isotopes in Hoba was undertaken to obtain both a terrestrial age and better information on the preterrestrial size and cosmic-ray exposure age. For comparison, the Cl^{36} and rare-gas isotopes were measured in Deelfontein, as were the Cl^{36} and Ni^{59} in Sikhote-Alin, the latter because it is a large iron meteorite of dated fall. The Ni^{59} is important as a neutron-produced isotope that could be used for evaluating the neutron effects of solar flares.

The terrestrial age of Hoba is a useful tool in the interpretation of the geologic and climatic history of the region. Postdating the fall of Hoba is a limestone deposit that has been laid over the surrounding granite. The 30 cm thick layer of unusual iron shale on the sides and bottom of Hoba is of related interest.

The preterrestrial size of Hoba is important because Hoba is the largest known meteorite and probably did not fragment in its atmospheric passage. Hoba contains no cracks nor planes of weakness, and, since it is a high-nickel ataxite, it is exceedingly uniform and does not easily fragment nor erode. Ablation from a large, uniform, nonfragmenting meteoroid is more susceptible to calculation than it is from other types of meteoroids.

The sample of Hoba used in this work was donated by Dr. and Mrs. Laurence Marshall. Dr. Charles E. Stott, General Manager of the Tsumeb Corp. of South-West Africa, kindly cut the sample. Mr. C. de Jager of South-West Africa gave us the sample of Deelfontein, and Dr. E. L. Krinov of the Academy of Sciences of the USSR donated the sample of Sikhote-Alin used in this work. We are grateful to them.

II. CHEMICAL ANALYSIS OF HOBA

For the analysis of Hoba, a 12 g piece adjacent to that used in the isotope study was freed of oxide crust and dissolved in nitric and hydrochloric acids. Iron was then determined by dichromate titration. Nickel was separated by dimethylglyoxime precipitation, redissolved, and titrated with EDTA by means of a murexide indicator (Welcher 1957). Cobalt was estimated by colorimetry by use of thiocyanate and acetone (Sandell 1956). The analysis yielded the following results: 82.34% Fe, 16.44% Ni, 0.78% Co.

III. CHEMICAL SEPARATION AND COUNTING OF RADIOISOTOPES

After the oxide crust was removed, the sample was weighed, carriers were added, and the sample was dissolved in acid. The Hoba and Deelfontein samples were dissolved in nitric acid, and the Sikhote-Alin sample in sulfuric acid. Silver chloride was precipitated from the resulting solutions. The silver chloride was purified several times by dissolving in ammonia, filtering the ammoniacal solution, and reprecipitating the silver chloride. The purified silver chloride was treated with H_2 gas at 600°C to produce HCl, which was converted to NH_4Cl, which in turn was weighed, and then counted under a low-level, thin-window β counter. The Cl^{36} counting results are given in Table I.

Iron was extracted with isopropyl ether, then back-extracted into water and stored. Cobalt and the remaining iron were removed from the raffinate onto an anion-exchange resin. The Co and Fe were eluted, and the Fe was removed from the Co by extraction with isopropyl ether. The Co concentration was determined colorimetrically; the cobalt was then precipitated as the oxide, and counted for Co^{60} by the γ-γ coincidence method with a pair of NaI (Tl) crystals 3 inches in diameter by 3 inches thick in a low-level shield. The Co^{60} results are given in Table II.

After the addition of 100 mg of Fe carrier, the Mn, Be, Al, Fe, and part of the Ni were precipitated with NH_4OH and H_2O_2. Nickel was precipitated from portions of the supernatent from this precipitation with dimethylgloyoxime. Nickel was leached from the precipitate with dilute HNO_3, and the resulting solution was taken to fumes with H_2SO_4. The Ni concentration was determined, and Ni was electroplated from aliquots of an alkaline solution onto 60 × 23 cm copper sheets and placed in a proportional counter for counting the x-rays from Ni^{59}. For Hoba, 8.32 g of Ni was plated, and for Sikhote-Alin, 3.3 g. A calibrated solution of Ni^{59} was obtained from Isotopes Inc., and 8.3 g of Ni and 3.3 g of Ni containing known amounts of Ni^{59} were plated onto copper sheets in a fashion identical to that done with the meteorite sample. Copper sheets containing blank Ni were also prepared and counted. Table III gives the Ni^{59} results.

The precipitate of Mn, Be, and Al was leached with 10 M NaOH, redissolved, and reprecipitated with excess NaOH and H_2O_2. The solution contains Al and Be; the precipitate, chiefly Mn. The precipitate was redissolved with HNO_3 + H_2O_2 and the Mn recovered and purified by repeated precipitation of it as MnO_2 with $NaBrO_3$. The final MnO_2 precipitate was ignited to Mn_3O_4, weighed, and counted with a thin-window proportional counter. The counting efficiency was determined by counting a known activity of Fe^{55} in Mn_3O_4 geometry.

TABLE I. Cl^{36}

Sample wt g	NH_4Cl counted mg	Chemical yield %	Activity cpm
Hoba (1465)	149.3	65	0.250 ± 0.001
Sikhote-Alin (73.8)	134.4	96	0.111 ± 0.002
Deelfontein (259.3)	121	61	0.471 ± 0.006

TABLE II. Be^{10}, Al^{26}, Mn^{53}, and

Isotope	Element carrier mg	Material counted form and wt. mg	Chemical yield %
Be^{10}	72	BeO (127)	63.5
Be^{10} (recycled)	72	BeO (60.9)	30.5
Al^{26}	100	Al_2O_3 (164)	87
Mn^{53}	20	Mn_3O_4 (21)	76
Co^{60}	(0.78%)*	CoO (10,450)	72

*Concentration in Meteorite

TABLE III. Ni^{59}

Sample	Ni content %	Ni counted g	6.9 kev x-ray ct/min— background
Hoba	16.44	8.32	0.09 ± 0.02
Sikhote-Alin	5.8	3.29	0.18 ± 0.03

The Al and Be were recovered from the NaOH solution by acidifying and precipitating with ammonia. The precipitate was then dissolved in HCl, and aluminum was precipitated with 8-quinolinol, ignited to Al_2O_3, weighed, and counted between a pair of NaI (Tl) crystals by the γ-γ coincidence method.

Counting Results.

Background cpm	Counter efficiency	Cl^{36} dpm	Cl^{36} dpm/kg
0.072 ± 0.002	0.120	2.29 ± 0.11	1.56 ± 0.08
0.060 ± 0.002	0.184	2.9 ± 0.30	3.9 ± 0.4
0.031 ± 0.002	0.178	4.1 ± 0.3	15.8 ± 1.2

Co^{60} Counting Results in Hoba (1465 g).

Ct/min background	Counter efficiency %	Activity dpm	Activity dpm/kg
0.038 ± 0.008	6.0	1.0 ± 0.2	0.68 ± 0.14
0.024 ± 0.009	7.4	1.06 ± 0.42	0.72 ± 0.28
0.009 ± 0.003	2.6	0.40 ± 0.14	0.27 ± 0.10
0.098 ± 0.005	0.084	154 ± 15	105 ± 11
0.020 ± 0.002	0.37	7.5 ± 0.8	5.1 ± 0.6

Counting Results.

Counter efficiency %	Ni^{59} dpm	Ni^{59} dpm/kg
0.50 ± 0.05	18 ± 5	350 ± 100
1.1 ± 0.1	16 ± 4	285 ± 70

The Be was precipitated as the hydroxide from the supernatent from the Al precipitation. It was ignited to the oxide and redissolved in a syrup of $(NH_4)SO_4$ and H_2SO_4. The Be was then purified by repeated precipitation of it as the hydroxide in the presence of EDTA. Nalgene labware was used. The final precipitate was ignited, weighed,

and counted with a low-level, thin-window β counter. Blank BeO prepared in the same way showed no β activity. The Al^{26} and Be^{10} results are given in Table II.

IV. RARE GASES

The rare gases were measured in a metal mass spectrometer with a radius of 6 inches. The gas extraction, purification, and calibration systems have been described by Fireman and DeFelice (1968). Aliquots of the gases from a 5 g sample of Hoba were analyzed. The He^3 content was $(25 \pm 3) \times 10^{-8}$ cc/g with a He^3/He^4 ratio of 0.215 ± 0.010; the Ne^{21} content was $(0.29 \pm 0.3) \times 10^{-8}$ cc/g with Ne^{20}/Ne^{21} and Ne^{22}/Ne^{21} ratios of 1.20 ± 0.04 and 1.06 ± 0.02, respectively. The Ar^{38} content was $(1.56 \pm 0.10) \times 10^{-8}$ cc/g with an Ar^{36}/Ar^{38} ratio of 0.62 ± 0.01. Two samples of Deelfontein were analyzed. The He^3 content was $(383 \pm 15) \times 10^{-8}$ cc/g with a He^3/He^4 ratio of 0.255 ± 0.005; the Ne^{21} content was $(4.9 \pm 0.3) \times 10^{-8}$ cc/g with Ne^{20}/Ne^{21} and Ne^{22}/Ne^{21} ratios of 1.03 ± 0.01 and 1.05 ± 0.01, respectively; the Ar^{38} content was $(24.2 \pm 0.6) \times 10^{-8}$ cc/g with an Ar^{36}/Ar^{38} ratio of 0.636 ± 0.005.

V. AMOUNT OF SHIELDING

The Cl^{36} in Hoba is 1.56 ± 0.08 dpm/kg; in Sikhote-Alin, 3.9 ± 0.4 dpm/kg; and in Deelfontein, 15.8 ± 1.2 dpm/kg. The low Cl^{36} activity in Hoba cannot be ascribed to decay since time of fall because Ni^{59}, which has a much shorter half-life than the 300,000 year half-life of Cl^{36}, is present. We must ascribe the low Cl^{36} to the shielding that surrounded the sample in space. The amount of Cl^{36} expected as a function of depth in iron spheres of various radii has been calculated by Kohman and Bender (1968). If the present mass of Hoba is approximated by a sphere, a radius of 180 cm is obtained. The Cl^{36} activity of 1.5 dpm/kg would be expected at a depth of 35 cm in an infinite body. If the size of the Hoba meteoroid is assumed to be that of the present meteorite, the Cl^{36} activity of 1.5 dpm/kg corresponds to a depth of 45 cm. Therefore, the depth of the Hoba sample corresponding to its Cl^{36} activity is between 35 and 45 cm. The Mn^{53} activity of 105 ± 11 dpm/kg according to these same calculations corresponds to a depth of 25 cm for an infinite body, and 40 cm for the present-sized body. A depth between 35 and 40 cm is consistent with both the Cl^{36} and Mn^{53} activities. At this range of depths, the Al^{26} and Be^{10} activities should be equal and have a value between 0.25 dpm/kg and 0.60 dpm/kg. The resultant values of 0.27 ± 0.10 dpm/kg for Al^{26} and 0.68 ± 0.14 dpm/kg for Be^{10} confirm the 35 to 40 cm

depth. Our sample was unoxidized metal. If a 15 cm thick layer of metal oxidized to form the observed 30 cm thick crust, the side from which our sample was taken lost only 20 to 25 cm by ablation.

The Cl^{36} of 3.9 ± 0.4 dpm/kg in our Sikhote-Alin sample corresponds to the activity expected at the center of a sphere 56 cm in radius; Cl^{36} measured in other samples of Sikhote-Alin ranges from 7.4 to 12.5 dpm/kg (Kaye 1963). All the observed Cl^{36} activities are possible in a sphere of 56 cm radius; the sample containing 12.5 dpm/kg would be at the surface, and other samples at various depths. The preatmospheric radius of Sikhote-Alin appears to have been close to 56 cm. According to the calculations of Kohman and Bender, the Cl^{36} activity of 15.8 dpm/kg in Deelfontein implies that the original radius of Deelfontein was between 25 and 35 cm. In fact, the recovered mass of Deelfontein was 28 kg, and it had dimensions of 20 to 30 cm (Comerford, McCorkell, and Tishler 1968). Our measurements indicate that the "larger mass" from which it is supposed to have come probably does not exist.

VI. TERRESTRIAL AGE

The nuclide with the shortest half-life (80,000 years) that we measured in Hoba was Ni^{59}. The Ni^{59} activity is 355 ± 100 dpm/kg. This is higher than the Ni^{59} activities measured in other iron meteorites, which range from less than 8 dpm/kg for Sardis to 133 ± 8 dpm/kg for a sample of Sikhote-Alin (Kaye 1963). (Our Sikhote-Alin sample yielded 285 ± 70 dpm/kg.) The maximum Ni^{59} activity expected from the action of cosmic rays in a meteorite with 5.8% Ni is 75 ± 25 dpm/kg; this value occurs at the center of a 70 to 80 cm radius sphere (Kornblum 1968). Our Sikhote-Alin sample came from approximately the center of a 56 cm radius sphere and has more than twice the expected Ni^{59} activity. The excess Ni^{59} activity may arise from the action of solar flares. Solar flares should contribute to the neutrons but not to spallation. The Ni^{59} activity is proportional to the Ni content; the Ni content of Hoba is 16.44%. The location of our sample in Hoba, 35 to 40 cm from the surface of a large body, corresponds to a Ni^{59} activity of 70 ± 20 dpm/kg from the action of cosmic rays, according to the calculations of Kornblum. The Ni^{59} activity of 350 ± 100 dpm/kg for the Hoba sample is more than twice the expected value. This situation is analogous to that in Sikhote-Alin, which is a recent fall, and leads to the conclusion that Hoba is also a recent fall in the sense that its terrestrial age is less than one half-life of Ni^{59} or 80,000 years.

Geological and climatic changes in the region in which Hoba is found can be deduced from the geological conditions in which the meteorite lies. The terrestrial age of Hoba requires these geological

and climatic changes to be of recent origin. Hoba does not lie in a crater, and there is no geological evidence of impact. Since its fall, it has been covered by a layer of calcareous tuffa or surface limestone that overlies granite (Spencer 1932). This probably formed by the evaporation of stagnant surface water bearing calcium carbonate from the nearby Otavi dolomite. The region today is arid, with an annual rainfall of only 20 to 30 inches and little runoff. Thus, it seems probable that a period of moister climate occurred in the region during the last 80,000 years, probably at the time of the last pluvial and glacial period in the Northern Hemisphere.

The oxidation crust or "iron shale" that covers the bottom and sides of Hoba contains Fe, Ni, and Co in the same proportions as does the metal (Spencer 1932), indicating that the oxidation occurred with no leaching. The oxidation probably took place rapidly during the moist period in which the surface limestone formed.

VII. EXPOSURE AGE

The Cl^{36} content of Hoba of 1.56 ± 0.08 dpm/kg can be combined with its Ar^{36} content of $(0.97 \pm 0.10) \times 10^{-8}$ cc/g to give a Cl^{36}-Ar^{36} exposure age. Since 82% of the Ar^{36} atoms arise from Cl^{36} decays, this exposure age is $(263 \pm 40) \times 10^6$ years. Within the limits of error, this exposure age is the same as the K^{40}-K^{41} exposure age of $(300 \pm 100) \times 10^6$ years (Voshage 1967). Space erosion would cause the Cl^{36}-Ar^{36} age to be younger than the K^{40}-K^{41} age; however, the errors are such that this difference is not apparent. Since Hoba is a nickel-rich ataxite, it would be the toughest type of iron meteorite to erode. The oldest K^{40}-K^{41} exposure age for any iron meteorite, 2.28×10^9 years, is for the nickel-rich ataxite Deep Springs. This is consistent with the idea that nickel-rich ataxites have the lowest rate of space erosion, less than 10^{-8} cm/year. Although Deep Springs is listed in Hey's catalog as a probable dated fall, we found less than 0.2 dpm/kg of Ar^{39} in it, an indication that it landed more than 1400 years ago. If a Cl^{36}-Ar^{36} age for Deep Springs is calculated by use of the value of 6.0 ± 0.6 dpm/kg for Cl^{36} (Kay 1963) and the value of 58.5×10^{-8} cc/g for Ar^{36} (Signer and Nier 1962), the anomalously high exposure age of 4.1×10^9 years is obtained. This leads to the suggestion that its Cl^{36} activity is too low, owing to decay since the time of fall. Since the Cl^{36} half-life is 300,000 years, Deep Springs landed more than 300,000 years ago. Therefore, Deep Springs has withstood the effects of weathering for a much longer time than has Hoba.

The Cl^{36} content of Deelfontein, a coarse octahedrite, is 15.8 ± 1.0 dpm/kg (see Table I). This Cl^{36} content can be combined with its Ar^{36} content of $(15.2 \pm 0.6) \times 10^{-8}$ cc/g to give an exposure age of

$(400 \pm 40) \times 10^6$ years. This is not an unusual exposure age for a coarse octahedrite.

VIII. Fe^{60} IN HOBA

Because Co^{60} has a half-life of only 5.27 years, its presence would not be expected in a meteorite such as Hoba West, except as the daughter of Fe^{60} or as the product of the capture of cosmic-ray neutrons by Co^{59} in the meteorite at the earth's surface. The Co^{60} from meteorite Fe^{60} has been measured only once, by Goel and Honda (1965); compared with their value, the Co^{60} activity in Hoba is high. Calculations using the methods and data of Montgomery and Tobey (1949), with consideration of resonance neutron capture, indicate that most or perhaps all the Co^{60} found in Hoba arises from the capture of cosmic-ray neutrons in meteoritic cobalt at the earth's surface.

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