Cosmogenic Rare Gas Contents of
IRON METEORITES

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". . . A stone of great size did fall, according to the common belief, from heaven, at Aegos Potami, which is shown to this day, and held in great esteem. . . ."

SO reads in part what is one of the earliest descriptions [1] of a meteorite fall. The account continues with a further description of the fall, along with possible explanations of the origin of the stone. Throughout antiquity and during the middle ages there were various reports of stones from heaven. The stones were preserved in museums and religious places. For, after all, a stone from heaven must surely be the evidence of some great force in nature. With the enlightenment of the Renaissance and especially with the cold light of scientific inquiry of the seventeenth and eighteenth centuries, meteorites were relegated to the ignominy of religious superstition.

A number of the falls, however, were reliably witnessed and the reports were well authenticated. Finally a German physicist, E. F. F. Chladni, was convinced of the trustworthiness of the accounts and published a paper [2] in 1794 assembling most of the evidence. The French Academy, then the highest authority in scientific matters, was finally convinced too in 1803 when a shower of 2000 to 3000 stones fell outside Paris at L'Aigle. The year 1803 then roughly marks the time when modern scientific methods were applied to meteorites. Meteorites were recognized as what is perhaps the only material on earth which is extraterrestrial in origin.

Today there are several thousand meteorite specimens in museums. This represents over a hundred years of collecting. It might be thought that a meteorite fall is

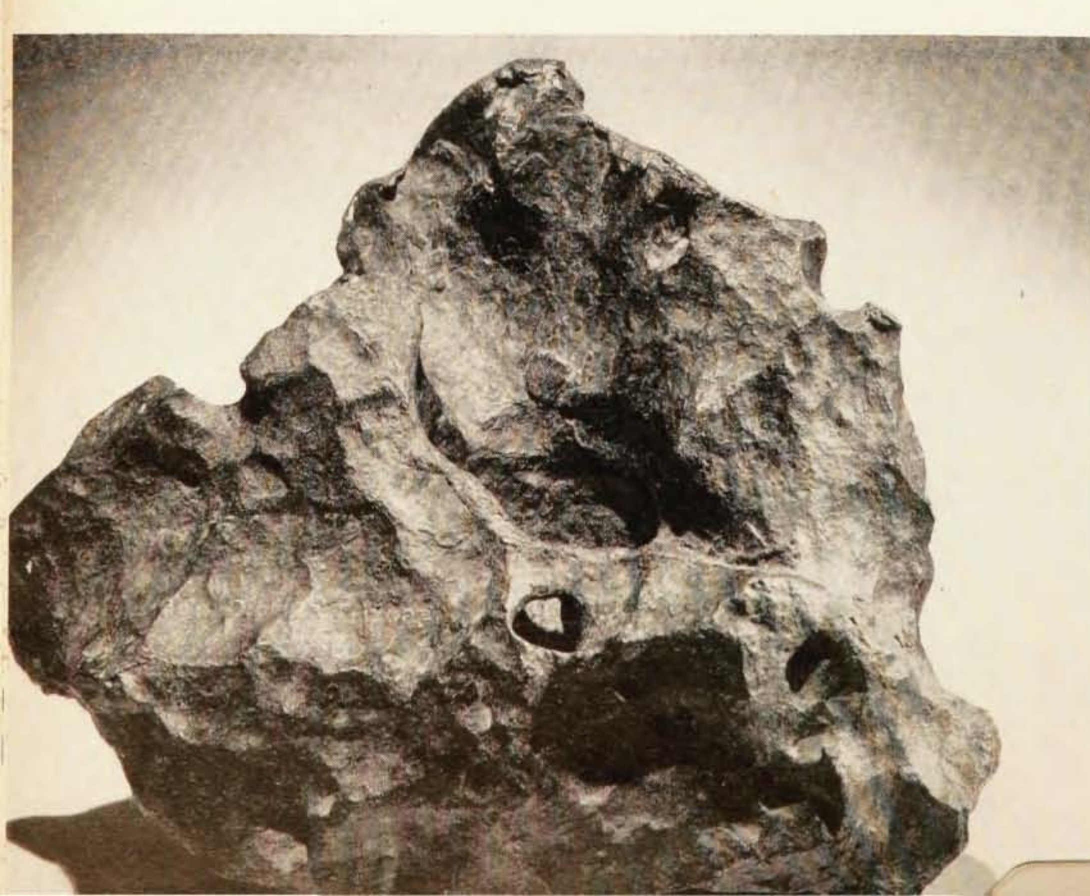

Fig. 1. A 95-pound piece of the Canyon Diablo meteorite. The fragment is roughly one foot in diameter.

a rare occurrence; actually such is not the case. It has been estimated that each day one thousand tons of meteoritic material are swept up by the earth's gravitational field. One-half ton is in the form of meteorites; the rest is extremely small particles which either vaporize on striking the atmosphere (meteors) or end up as dust particles of the upper atmosphere (micrometeorites).

Meteorites fall into three classes: (1) those which are almost a pure nickel-iron alloy, the iron meteorites; (2) the other large class of meteorites, the stones; and (3) a smaller class, which consists of a mixture of stone and iron phases, the stony-irons. Meteorites obtain their irregular surface from damage during passage through the atmosphere. Fig. 1 shows a 95-pound specimen of the Canyon Diablo iron fall. Iron meteorites show a characteristic crystal pattern on etching, the Widmanstätten pattern, shown in Figs. 2 and 3. The lamella can be wide, as in the Sikhote-Alin meteorite, or narrower, as in the meteorite Williamstown.

Meteorites furnish our only sample of extraterrestrial material and as such (at least until man is freed from his earthly prison) are the only means of studying material from other parts of the solar system and perhaps other parts of the galaxy. In this way, meteorites are likely to contain clues as to the origin of the solar system.

One theory as to the origin of meteorites is that they resulted from the break-up of a planet of a size somewhat smaller than the earth but large enough to have had a nickel-iron core. The iron meteorites then could be from the core of the protoplanet, while the stony meteorites could be from the mantle. The iron meteorites could be samples of the solar system which solidified early in the evolution of the planets, in contrast to the surface rocks of the earth which date from much later recrystallizations. On the other hand, meteorites may be remnants of the material from which the planets are made. In either event, iron meteorites may be the oldest minerals available on the earth.

A study of the age of iron meteorites should be very interesting in understanding the age of the solar system. To measure the age of an object one needs to measure the concentration of a long-lived radioactive isotope and its decay product. With a knowledge of the half life it is then a simple matter to calculate an age. For an object expected to approach the age of the solar system one needs a radioactive isotope with a half life of a billion years or so—long enough so that a measurable amount of it is still present. The radioactive chains starting from uranium and thorium produce different amounts of helium from the alpha emitters. Uranium, thorium, and helium were measured in a number of meteorites.[3] On computing the ages of the meteorites it was found that some were considerably older than anyone would seriously propose for the age of the solar system.

To understand this apparent paradox it was suggested[4, 5] that the helium may possibly be produced in meteorites by the action of cosmic rays. It was known through the study of irradiated photographic plates that cosmic rays produce nuclear reactions and that helium is one of the products. If this were the case, it was reasoned that the helium content should be less on the inside of an iron meteorite, which is more shielded from cosmic rays, than the outside, which is closer to the original surface in space. On the other hand, if the helium comes from the decay of uranium and thorium, which are more or less uniformly distributed through the meteorite, the helium should also be uniformly distributed. To check this point the depth variation of the helium content of several meteorites was determined.[6] On going through 20 cm of iron, the helium concentration was found to decrease only slightly, though by the

Fig. 2. A piece of the Sikhote-Alin meteorite.

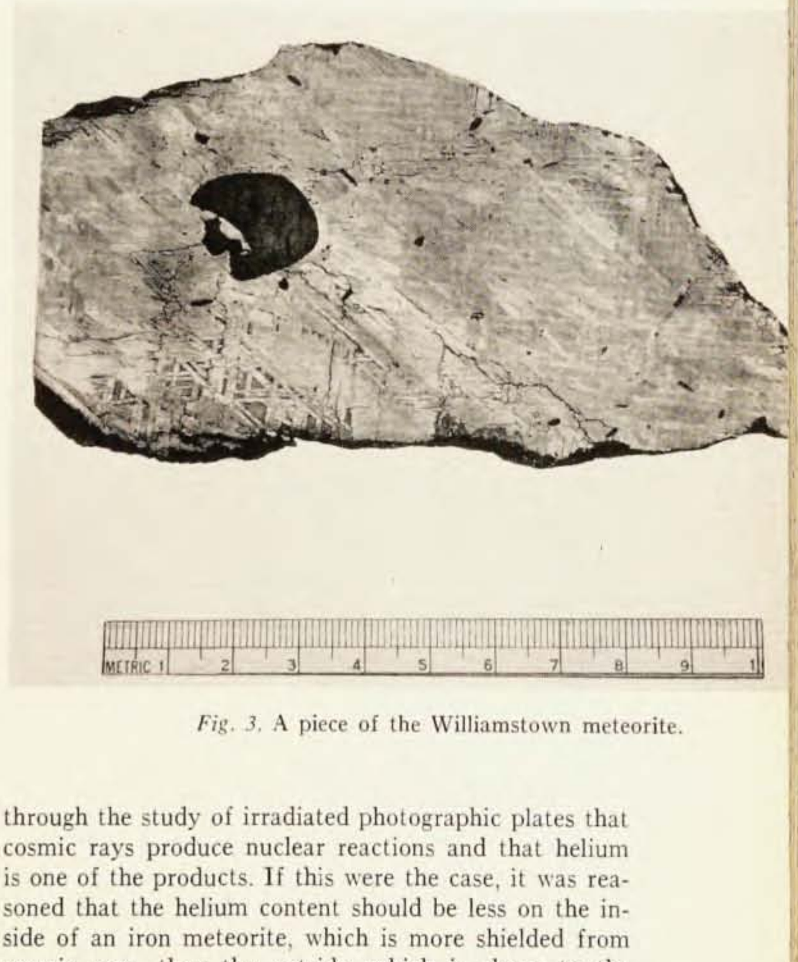

Fig. 3. A piece of the Williamstown meteorite.

then prevalent notions of the method of production of helium from cosmic rays, the decrease should have been considerable.

Another test was available. There are two isotopes of helium: helium-4 and helium-3. All the helium from uranium and thorium is definitely helium-4, while that produced by cosmic rays could contain appreciable helium-3. When a sample of meteoritic helium was analyzed isotopically,[7] it was found to contain about 25% helium-3. Terrestrial helium contains only approximately one part helium-3 for each million parts of helium-4. The large concentration of helium-3 is conclusive evidence that the helium is neither terrestrial helium nor helium from the decay of uranium and thorium. It has recently been found that helium is more uniformly distributed in the meteorite Carbo [8] than in another meteorite, Grant,[9] and it is suggested that possibly Carbo had been heated at some time.

If helium is produced by nuclear reactions induced by cosmic rays, then every other nuclide below iron should also be produced. It was found that neon and argon are also present, and recently potassium has been reported.[10, 11, 12] A number of radioactive isotopes have been found: [13, 14, 15, 16] among them are tritium, Ar^{39}, Cl^{36}, Al^{26}, Be^{10}, and Co^{60}.

By measuring the rare gas concentration and amounts of radioactive nuclides it is possible to have a probe on cosmic rays in other space and to learn about the constancy of cosmic rays in time and space. The study can lead to information important to an understanding of the origin of meteorites and possibly the solar system.

When an object such as a meteorite is traveling through outer space it is bathed in a flux of cosmic-ray particles. These high-energy particles break up the nuclei of the material and produce every nucleus lighter than those present in the meteorite. The stable nuclides build up in time at a rate depending on the local cosmic-ray flux. The radioactive nuclides increase and after being irradiated for several half lives they level off at a constant level so that the decay rate just balances the production rate. After the meteorite falls on the earth the production is considerably lowered because of the lower cosmic-ray flux on the earth's surface than in space. The stable elements remain essentially the same as the day the meteorite fell, while the radioactive nuclides decay.

The steady-state concentration of the radioactive nuclides is proportional to the production cross section and the cosmic-ray flux. If the cosmic-ray flux changes, the steady-state concentrations gradually drift to new values. The shorter the half life, the quicker radioactivity reaches its new value. On the other hand, the activity level of an isotope like potassium-40, with a half life of 1.3 billion years, responds only slowly to a change in cosmic rays. By measuring the activity levels for two separate isotopes of differing half lives and also measuring the production cross section, using high-energy accelerator-produced protons, it is possible to determine the variation in the intensity of cosmic rays in time. Such an investigation [13] has been made by studying the isotopes Cl^{36} and Ar^{39}. From the proton production cross sections the production ratio of Cl^{36} to Ar^{39} should be 1.0 ± 0.2. The observed ratio of these two isotopes in the Sikhote-Alin meteorite which fell in 1947 is 1.0 ± 0.1. It is concluded from this experiment that the intensity of cosmic rays has been constant, within 10%, for the last 100 000 years. By studying even longer-lived activities the possibility exists for testing whether the flux of cosmic rays has been constant for billions of years.

When a large iron meteorite is in space the portion near the surface is more intensely irradiated than the portion near the center, which is shielded by the iron surrounding it. As mentioned previously, it has been found that the intensity drops off. It drops exponentially with a coefficient of about 500 grams/cm², so that for a given fragment the amount of a given nuclide depends on the depth in the original meteorite and also on the time it has spent in space. On the other hand, a ratio of isotopes as He^3/He^4 should be independent of the age and depend only on the depth. If a scale of depth vs. isotopic ratio could be established, one would have a means of determining the times meteorites have been in space.

To make the required measurements one needs a sensitive mass spectrometer and a sample handling technique which will remove the rare gas from the target and not contaminate it with natural abundance material.

A mass spectrometer which is capable of detecting less than a million atoms of a rare gas is shown in Fig. 4. The analyzer tube is all glass, coated on the inside with tin oxide to be conducting. The tube is mounted in such a way that the magnet can be lowered and an oven lowered over the tube for baking at 450°C. In this way it is possible to obtain a vacuum of better than 10^{-10} mm of mercury. The spectrometer is oper-

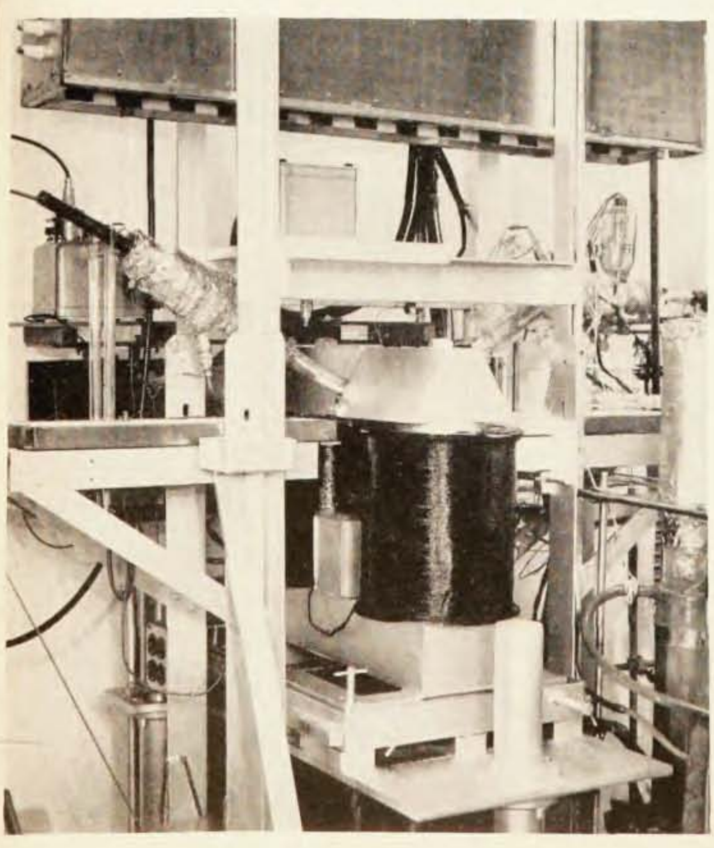

Fig. 4. Mass spectrometer for determination of trace amounts of the rare gases.

ated under static conditions; that is, the diffusion pump is isolated when the sample is admitted to the tube. This method of operation permits the detection of less than a million rare gas atoms. However, it is not feasible to operate with such small samples, as the amount of gas added during the preparation of the sample is many orders of magnitude larger.

As important as the mass spectrometer is the sample preparation system. The crucial problem in measuring small rare gas samples is the amount of rare gas added from extraneous sources. This gas is always orders of magnitude larger than the sensitivity of the mass spectrometer. The problem is how to melt a piece of metal without seriously outgassing the vacuum walls and to prevent the diffusion of helium through the hot walls. Helium is notorious for its leakage through glass, and for this reason presents a special problem. For example, if the walls are made of Pyrex, about ten times as much helium diffuses into the apparatus as is present in a typical sample. It has been found that glass which has alumina added to it is quite impervious to helium. Such a glass is made commercially for frying pans, since the added alumina increases the strain temperature. This glass (Corning formula 1720) is 1/10 000 times less porous to helium than ordinary Pyrex.[17] A sample line constructed from this glass controls the helium leakage problem.

An apparatus suitable for sample preparation is shown in Fig. 5. A sample of an iron meteorite or a piece of iron bombarded by high-energy protons is slowly vaporized out of an aluminum crucible heated by a surrounding molybdenum cup with rf induction heating. Impurity gases originating from oxygen, hydrogen, and carbon in the metal are formed by the high temperature. Water, hydrogen, and various organic compounds are produced and are removed by hot calcium and copper oxide. After the sample is prepared, it is expanded into the mass spectrometer and measured. A standard amount of the rare gases is then added by a gas pipette and the sample is measured again. In this way an accuracy of $\pm$ 5% is obtainable.

By this procedure it has been possible to measure the production cross sections for the rare gases from proton bombarded targets. The technique is capable at present of measuring amounts of helium, neon, and argon as low as 10^{-9}, 10^{-10}, and 10^{-9} cc, respectively, at NTP. To compare the proton production cross sections to the rare gases in meteorites, one must compare the $\sigma_{He^3} + \sigma_T/\sigma_{He^4}$ to the He^3/He^4 ratio in meteorites as the tritium decays to He^3. In Fig. 6 is plotted the ratio $\sigma_{He^3} + \sigma_T/\sigma_{He^4}$ as a function of energy. As can be seen, this ratio increases continuously up to 6 Bev. One then expects the He^3/He^4 ratio to drop on going from the surface to the center of a meteorite. This has been observed in the Grant meteorite.[9] The He^3/He^4 ratio decreases by about 10% in going from the surface to the center. Since the Ne and Ar production cross sections are also fairly energy independent in the Bev energy range, one would expect a similar behavior for the ratios of He^3 to neon or argon. Thus if one samples from the outside to the inside of a meteorite, the drop in the ratios is due to a depth energy dependence. The decrease may be interpreted as a change in the ratio of high-energy cosmic-ray protons relative to secondary protons, neutrons, and mesons. It should be possible to construct a scale of He^3/He^4 ratios as a function of depth; or, similarly, ratios of He^3/Ne^{21} or He^3/Ar^{38} may be used.

Only in a few meteorites have all three of the rare gases helium, neon, and argon been measured. A list

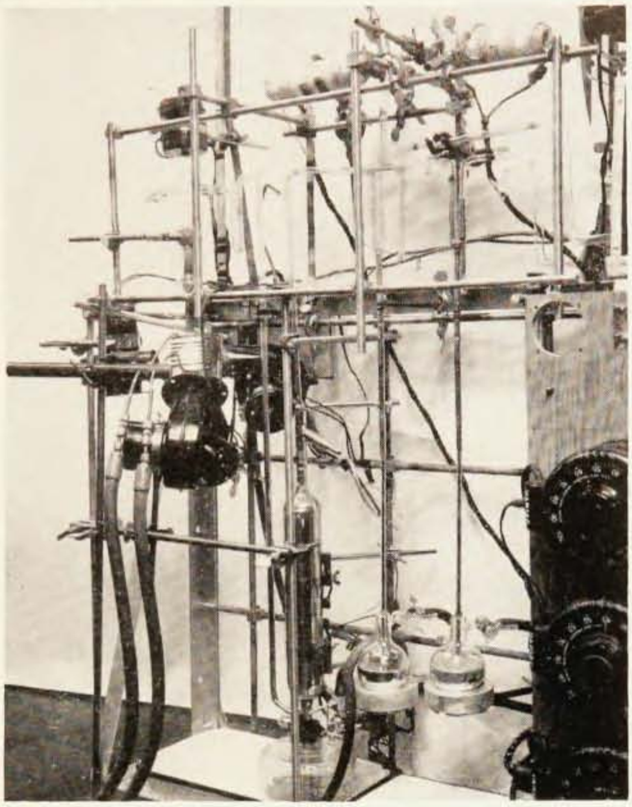

Fig. 5. System for the extraction of trace quantities of the rare gases from metals.

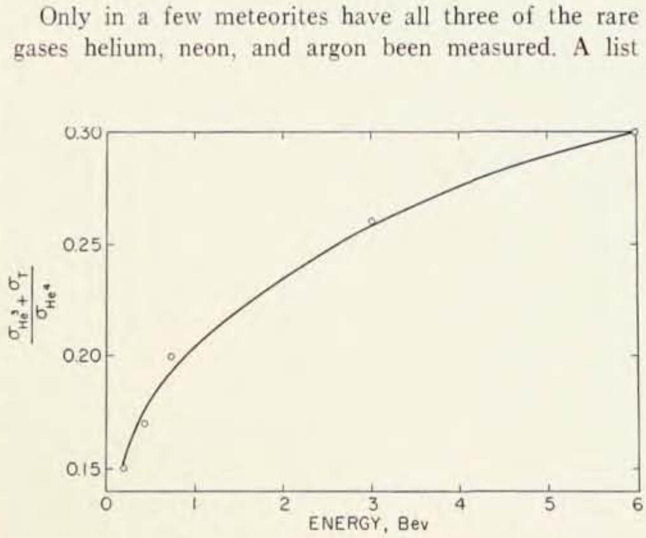

Fig. 6. The variation of helium and tritium proton production cross sections with energy.

Table I. Rare gases in iron meteorites

Meteorite	He^3/He^4	He^3/Ne^{21}	He^3/Ar^{38}
Casas Grande	0.35	130	36
Toluca	0.34	125	42
Arispe	0.29	117	30
Odessa	0.31	95	18
Sikhote-Alin	0.27	87	32
Williamstown	0.25	80	18

Table II. Exposure ages of iron meteorites

Meteorite	He^3 10^{-8} cc/g	He^3 *Age* (*millions of years*)	$Cl^{36}-Ar^{36}$ *Age* (*millions of years*)
Casas Grande	130	210	1000*
Toluca	100	170	—
Arispe	270	680	—
Odessa	220	550	400
Sikhote-Alin	130	310	270
Williamstown	480	2400	2100

* This age is probably high because some of the Cl^{36} activity has decayed since its fall.

of some of these is given in Table I. In general it is seen that the He^3/He^4, He^3/Ar^{38}, and He^3/Ne^{21} ratios all drop in value. In a case where a particular ratio is not in the usual order it may be possible that an inclusion may be contributing to one of the isotopes. For instance, an inclusion high in magnesium will contribute an excess of neon isotopes while a calcium inclusion will contribute an excess of argon isotopes. Uranium, if present, will contribute excess He^4. It may well be that when a meteorite shows agreement in ordering in all ratios but one, an impurity is the cause of the ratio being out of line. Such may be the case, for example, in the He^3/Ar^{38} ratio in the Odessa meteorite, in which a calcium impurity would be expected.

If one can put an actual scale on the depth (or equivalent production rate) vs. isotope it is possible, from a mass spectrometric measurement of the rare gas contents of an iron meteorite, to calculate the time the meteorite has been exposed to cosmic rays. The total content of helium or other rare gas is the integrated production. So if one had a method of measuring the rate one would have a simple way of obtaining the age. In principle, at least, the ratio of He^3/He^4, He^3/Ar^{38}, or He^3/Ne^{21} will determine the rate. One makes the tacit assumption, which can be verified experimentally, that cosmic rays are constant in time and space. The ratios will be dependent on the depth in the original meteorite in space. The only difficulty is in applying a scale to the plot of depth vs. ratio. This can be done by realizing that another way of obtaining the production rate is to measure the present activity level of a radioactive isotope having a half life which is short compared to the irradiation time (almost always over 50 million years), and which is long compared to the time since the meteorite has fallen. A good choice for this purpose is Cl^{36}, with a half life of 308 000 years. By measuring the Cl^{36} activity it is possible to arrive at a Cl^{36} production rate. Then from the ratios of the proton production cross sections one can obtain the production rates of the rare gas isotopes. Using this scale, exposure ages for the other meteorites can be calculated. This is shown in Table II. Also listed in the table are the Cl^{36}-Ar^{36} ages. The latter should be more reliable when available, as 80% of the Ar^{36} comes from the decay of Cl^{36}.

The ratio of $\sigma_{He^3} + \sigma_T/\sigma_{Cl^{36}}$ needed to compute the He^3 ages from the relation between He^3 production and the He^3/He^4 ratio from the He^3 content was obtained by using values for the cross section at 6 Bev and the ratios of the rare gases in the meteorites.

The large spread in ages of these results is not what might be expected from some of the current ideas on the origin of meteorites. The large spread in ages is consistent with the origin of meteorites from the astroidal belt. The astroidal belt contains many planetesimals which have a small but finite probability of colliding with each other. It is possible that such collisions expose material buried in larger pieces to cosmic rays. The spread in exposure ages would then be a characteristic of the collision rate of the asteroids. It is quite likely that by further studies of the rare gases in iron meteorites a history of meteorites can be pieced together. One can see the possibility of obtaining good estimates of the original age of meteorites before entering the earth's atmosphere, as well as the times the meteorites have been in space and on the earth.

So that in the words of Akenside:

Say from what simple springs began
The vast ambitious thoughts of man,
Which range beyond controul,
Which seek eternity to trace,
Dive thro' th' infinity of space,
And strain to grasp the whole?

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