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Cosmogenic Nuclides Under Shielding in Meteorites

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Abstract

Products induced in meteorites in space caused by interactions between cosmic rays and meteoritic materials are effected by shielding. Quantitative measures of the shielding are important in reliable estimations for production rates of cosmogenic nuclides and irradiation histories of meteorites. Irradiation histories are those of meteorites since born in space as meteorite. To describe the shielding in general, two independent variables are necessary, which are corresponding to depth of sample and size of meteorite. Conventional noble gas indexes found in chondrites have been compared with those of general terms of shielding, and an illustration of these indexes plotted in a unified diagram is proposed.

1. Introduction

The first study on cosmogenic products has been initiated in 1952 by Paneth *et al.* They found about 30% of ^{3}He in He fraction extracted from iron meteorites. The presence of ^{3}He as cosmogenic products in meteorites were suggested before by cosmic-ray physicists. Paneth *et al* had been anxious to determine the formation age of irons from the pairs of radiogenic ^{4}He and uranium, for years.. The difficulty was to eliminate the contaminations of uranium in samples and laboratory at the levels of ppbU. It may be surprizing to learn that even after their discovery of ^{3}He, very low He contents found in some large irons were attributed, in error, to extremely short formation ages of the irons.

Besides ^{3}He, many others such as ^{4}He, Ne isotopes, ^{38}Ar, and ^{36}Ar have been identified as high energy nuclear reaction products caused by cosmic-ray irradiations. Their symmetrical spatial distributions in a spherical body of an iron meteorite, Grant, were determined by Hoffman and Nier (1958) for He isotopes, and Signer and Nier (1960) for Ne and Ar isotopes. The lowest contents of these nuclides have been found at the center of a cutting surface of the meteorite. The steepest contour pattern was found for Ne isotopes; the flatter one was for ^{4}He.

Absolute estimations for interactions between cosmic ray secondaries and thick meteoritic bodies are complicated problems and hard to perform in general cases. Fluxes, intensities and spectra, of secondaries originated from galactic cosmic-ray primaries of higher than 1 GeV/n (n: nucleon), and high energy reaction cross sections in a thick target have to be estimated unde various geometries and materials. The important farctions of cosmic-ray secondaries are high energy neutrons, and studies on the reactions by these neutrons cannot be easily performed. One method is to extend our knowledges for current atmospheric cosmic ray irradiations and for high energy spallation studies by accelerators. Another approach can also be extended more empirically applying a thick target, a model of meteorites, and bombarding them by a high energy proton flux.

2. Universal Measures of Shielding in Meteorites.

Relative production rates of many products in a wide range can be expressed by a simple power function of mass loss from iron target (Geiss *et al*, 1962; Stauffer and Honda, 1962). That is,

$$p(A,Z)=f\times k_1\times(\Delta A)^{-k_2}.$$

for a total isobaric yield from ^{20}Ne to ^{45}Sc, $f=1$ can be used. This simple relation is due to a statistical nature of the high energy spallation reaction occurred in some hundred mega electron volt regions. Geiss *et al* derived above relation assuming a simplified power function of energy for the flux inside meteorites. In the equation above, k_2 is indicating a hardness of cosmic ray flux. With higher k_2 lower energy products of small ΔA values are observed in higher yields. The k_1 is, on the other hand, reflecting the intensity of the flux which results a general production level. The k_1 and k_2 are both considered to be two independent measures of shielding effects. By the use of "shielding", we can estimate directly the production rates for various products in various meteorites. In this case, the shielding can be classified as a self shielding; it is due to nuclear sensitive materials between source of flux and the sample to be examined. A reduction or attenuation of flux intensity can be referred as in the iron meteorite Grant. In smaller meteorites or in shallower depths, it may also inform a higher multiplicity of secondary particles, that means a increase of productions of lower energy products with depths or size of meteorites. Extensive applications of above statistical relations can be made based on empirical facts. The range of products can be extended up to $\Delta A=2$, or $A=54$ such as ^{54}Mn, when we simply modify the ΔA value to $\Delta A'=\Delta A+$a (Imamura *et al*, 1980). This must be only by purely empirical reasons; a correction term, "a", can be assigned to 4 as the best fit. When we employ $\Delta A'$ instead of ΔA, k_1 and k_2 both are also slightly modified to k_1' and k_2' respectively. That is,

$$p(A,Z)=f\times k_1'\times(\Delta A')^{-k_2'}.$$

$\Delta A'$ and f values of various products so far determined are tabulated in Table 1.

For small size meteorites, such as usually available chondrites, k_1' can be approximately expressed with k_2',

$$k_1'=0.042\times(23)^{k_2'} \qquad \text{atom/min. g.}$$

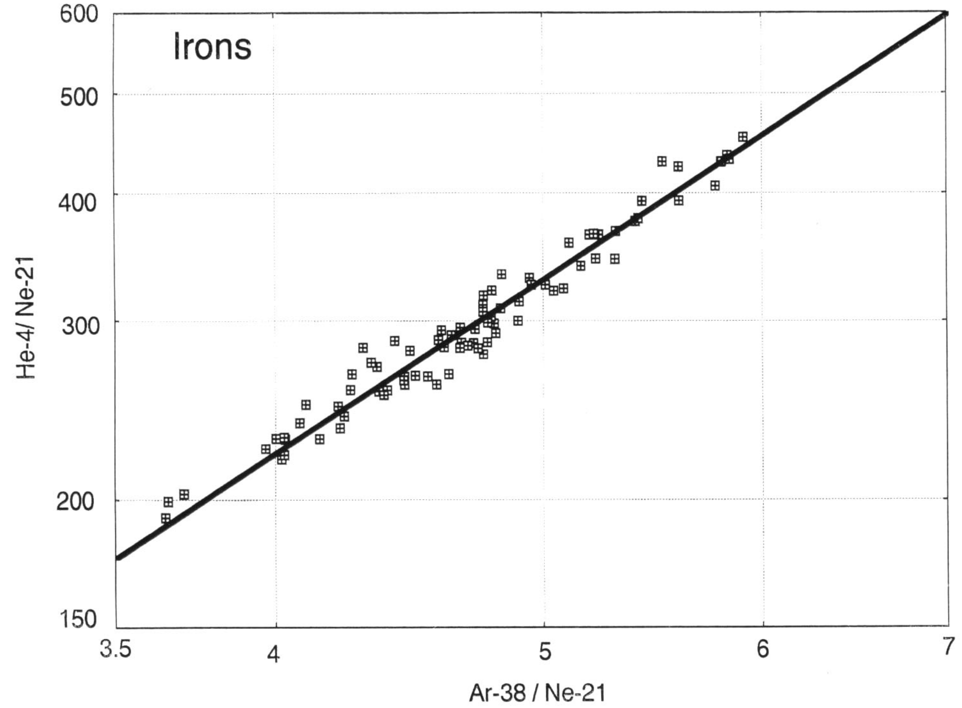

Fig. 1. Relation between $^4He/^{21}Ne$ and $^{38}Ar/^{21}Ne$ in iron meteorites. (based on data by Voshage *et al*).
The two sets of noble gas ratios are observed in a simple power function.

Therefor,

$$p = f \times 0.042 \times (\varDelta A'/23)^{-k_2'} \quad \text{atom/min. g.}$$

According to this equation, shielding can be expressed with a single variable, k_2', and production rates for product having $\varDelta A' = 23$ must be constant throughout a whole range of k_2'. Such examples, or the similar cases, are rather common; they are ^3He, ^{38}Ar, ^{10}Be, ^{36}Cl, ^{40}K and their neighbors in metal and also ^3He in a bulk chondrite. Those are also classified as higher energy products.

Based on spallation systematics, for lower mass products, $A < 20$, the simplification could not be expected. For a high yields of the smallest mass products, such as ^3He and ^4He, some special considerations seem to be necessary. They are not spallation products but classified to evaporation products which are resulted by lower energy process accompanying with high energy spallation. According to light noble gas data found in various iron meteorites, however, there seem to be another simple relations between their concentrations. Figs. 1 and 2, for examples, may indicate obviously such simple relations among their concentration ratios. Any set of the ratio can be directly compared with others in a power function. This simple fact

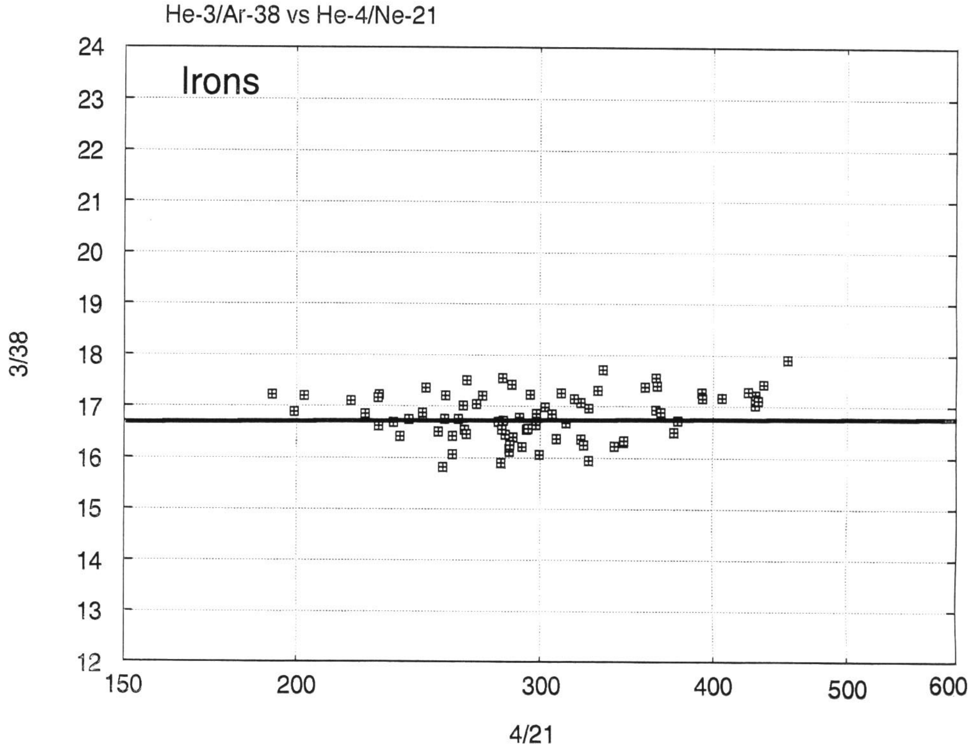

Fig. 2. Relation between $^4He/^{21}Ne$ and $^3He/^{38}Ar$ in iron meteorites. (based on data by Voshage *et al*).

^3He and ^{38}Ar are produced in a constant ratio, 17: 1 in iron meteorites. The ratios seem to increase slightly in both lower and higher shielding conditions of $k_2'=2.7$ or $^4He/^{21}Ne=300$.

was found among the data which have been intensively obtained by Voshage *et al* in 1978; 1979; 1980 and 1983. For example, $^3He/^4He$ are also compared with $^{38}Ar/^4He$ and a single monotonous smooth curve can be observed. According to our interpretation their logarithms can simply be plotted in a diagram and one straight line is obtained to correlate these ratios covering a whole range. That is exactly what we can expect from above k_1 and k_2 variables. In short, here also we may introduce equivalent equations for ^3He and ^4He just like on for ^{38}Ar and ^{21}Ne. Only thing we have to admit is to adopt some apparent $\Delta A'$ and large f values as shown in Table 1.

It may be instructive to learn that the systematics of He isotopes has been found after more than 30 years of the discovery. Besides, this finding of statistical behaviors of He encouraged us to extend the same idea to all other cosmogenic products in various samples of irons and stones.

The limits of applicabilities, however, must be examined carefully, because the

Table 1. Parameters used for Production Rate Estimation.

Target	Radioactive Product	$\Delta A'$	f	Stable+K40 Product	$\Delta A'$	f
Irons	Mn-54	6	0.59	Cr-54	6	0.6
	Mn-53	7	0.78	Cr-53	7	1
	Mn-52	8	0.35	V-50	10	0.49
				Cr-50	10	0.47
	V-49	11	0.86	Ti-50	10	0.03
	V-48	12	0.6	Ti-49	11	1
	Sc-46	14	0.42	Ti-46	14	0.98
				He-4	14	18.3
				Sc-45	15	1
	Sc-44m	16	0.2	Ca-44	16	1
	Ti-44	16	0.04			
				Ca-43	17	1
				Ca-42	18	1
	Ca-41	19	0.5	K-41	19	1
				K-40	20	0.64
	Ar-39	21	0.5	He-3	22	16.7
	Be-10	22	0.11	Ar-38	22	1
	Cl-36	24	0.64	Ar-36	24	0.78
	Al-26	34	0.23	Ne-21	39	1
L-chondrite	Al-26	7	0.10♯	Ne-21*		
	Na-22	9.5	0.33	Ne-22	9.5	0.74
	Be-10	11	0.10			
	Be-7	ca. 10	0.40	He-3	23	22
	C-14	ca. 8	0.12			
Carbon	Be-10	10	0.45			

*: current statistical method cannot be applied.
p(Ne-21)=p(Ne-22)/(Ne22/Ne21)

♯: According to observed data of Knyahinya, $f=0.11$ seems to be better.

For short lived nuclides, shorter than Na-22, solar cycle effects can be applied in some extents depending on the time of fall.

excitation functions for the He isotopes are substantially different from those of other typical spallation products. Their crosssections increase gradually in a wide energy range from 10 MeV to 10 GeV regions. For instance, the behaviors of ^{3}He relative to ^{38}Ar with shielding, depth and size effects in iron meteorites, look quite parallel in a wide range. That must not be true in a strict sense, but the ratios of ^{3}He/^{38}Ar, which are about 17, are only slightly higher, by a few %, in both lower and higher shielding regions than in medium size irons, at about $k_2'=2.7$ or ^{4}He/^{21}Ne=300. Only because changes of the spectra are in a wide range, and the total sum of multiplications of energy and cross-section terms is rather insensitive to the differences in the excitation functions. The flux, having a form of $(E_0+E)^{-2.5}$ can be employed varying $E_0=1000$—100 MeV. A similar treatment to ^{3}He and ^{38}Ar can also be applied for ^{10}Be, a fragmentation product, in metal. According to literature the excitation function for ^{10}Be production seems to have a reasonable intermediate pattern with those of ^{38}Ar and ^{3}He.

In any case, according to this relation our scope to systematizations becomes wider and more useful for many varieties of extra-terrestrial materials. As described above, the excitation functions for He isotope productions are not like those of simple spallation reactions which have different threshold energies, and beyond the thresholds the increase of spallation cross-sections levels off and become invariable with energy. The cross-sections for He isotopes seem to correspond to those of spallation reactions in a complex mixture of different mass targets. This consideration may lead us to apply the same technique to multi-element targets such as chondrites.

Production rates of ^{3}He and ^{22}Ne in chondrites can be estimated using parameters listed in the Table 1. On the other hand, those for ^{21}Ne cannot be given, because of a complicated production mechanism for ^{21}Ne, and because of an importance of the delicate behavior of ^{21}Ne relative to ^{22}Ne, as described in the following sentences. In general the most impressive features of the spallation products may be found in their extensive statistical behaviors. The examples can be seen in the equal productions of three Ne isotopes in irons as well as in stones in spite of a large difference in their $\Delta A'$ values. ^{3}He productions in irons and stones are also controlled by surprisingly similar set of parameters.

3. Shielding Indexes by Noble Gases in Chondrites.

For chondrites, the shielding has been expressed by empirical concentration ratios of ^{3}He/^{21}Ne, or simply 3/21, and ^{22}Ne/^{21}Ne, or simply 22/21. In 1966, Eberhardt *et al* reported an important relation among ^{3}He/^{21}Ne and ^{22}Ne/^{21}Ne in 30 species various chondrites, as

$$^{3}\mathrm{He}/^{21}\mathrm{Ne}=2.40+23.4\times(^{22}\mathrm{Ne}/^{21}\mathrm{Ne}-1)\,.$$

The relation can be quoted informally as "Bern line". The left term, ^{3}He/^{21}Ne, changes usually from 3 to 8; the higher ratios indicate the lower shielding. Similar relations have been proposed already, but they are all by simple linear expressions between the ratios. The latter, ^{22}Ne/^{21}Ne, varies only 20% or so; usually in a range of 1.05–1.25. The measurement, however, can be made with a higher accuracy for Ne isotope ratios, much better than ±1%, and there is much less possibility of diffusion loss by escape. Therefor ^{22}Ne/^{21}Ne has been taken to be more reliable as the shielding index. The production rate of ^{21}Ne can also be estimated from the Ne isotope ratio, and the irradiation, or exposure, age, a time of duration of cosmic irradiation to the meteorite, can be informed from ^{21}Ne. In fact, the absolute value of production rates as a function of shielding is not easily estimated correctly. For this purpose, we have to determine radioactive nuclides at the saturation level, as well as the stable isotope as the decay product. The same technique as iron meteorites can be used with metal fraction of chondrites. The age and then the rate can be calculated. Eugster (1988), for example, gave

$$p(^{3}\mathrm{He})_{L}=2.09-0.43\times(22/21)\quad 10^{-8}\ \mathrm{cc/g.\ my},\ \text{for L-chondrite.}$$

Above equation is indicating an essentially constant production rate of ^{3}He under various shielding in chondrites.

The mechanism to produce ^{22}Ne can be interpreted to be by spallation mainly in Mg and Si. For ^{21}Ne, however, two possibilities are considered; one is by spallation the same as that of ^{22}Ne, the others is by ^{24}Mg (n, α) ^{21}Ne. By spallation alone, ^{21}Ne/^{22}Ne must be lower with shielding, whereas by addition of (n, alpha) to ^{21}Ne, ^{21}Ne/^{22}Ne can increase instead, and that is what we found in chondrites. Unfortunately at this stage, this qualitative explanation is only all we can tell on the peculiar but useful behaviors of Ne isotopes.

However, the values of ^{3}He/^{21}Ne are not strictly corresponding to ^{22}Ne/^{21}Ne in an one-to-one correlation (Fig. 3). In other words, a shielding cannot be expressed uniquely by one set of the ratio, but the shielding can only be expressed in a range of about ±10% in the diagram of two sets of data. Actually the depth profiles of various chondrites are distributed in a set of parallel lines in a diagram, and the points for surface samples systematically locate in lower or right directions than those for near center samples. This situation seems to be similar to the relations of k_1' and k_2', at least qualitatively. For an accurate description of a shielding we need a two dimensional plot of two sets of noble gas data.

In fact, under usual geometries of chondrites, ^{3}He productions are not variable much, and ^{3}He/^{22}Ne is proportional to the reciprocal of the production rate of ^{22}Ne. On the other hand, ^{3}He/^{22}Ne seems also to indicate the degree of spectral shape of the flux, corresponding to the k_2'. The similarity between two sets of diagrams concerning shielding can be illustrated as in Figs. 3 and 4.

A serious risk to employ ^{22}Ne/^{21}Ne is in a different aspect; that is, as a measure of heavier shielding the ratio is loosing the sensitivity. For lower than 3 in the ratio of ^{3}He/^{21}Ne, we cannot figure out the real shielding by inspecting ^{22}Ne/^{21}Ne. Originally Ne ratios are proposed to be a measure for shielding for ordinary size chondrites. Even among chondrites larger objects have been studied and to these materials there is no good convenient measure for shielding from noble gas data. Some may be indicated by lower ^{3}He/^{21}Ne, but the lower values cannot be distinguished with loss of ^{3}He during irradiation in space. In this situation, sets of radio nuclides have to be studied at the levels of reproducibility of noble gases.

In general, stable and radioactive pairs must be studied simulaneously equally well, and a consistent result between the two can only guarantee a realistic picture of irradiation history of meteorite. Otherwise possibilities of interferences cannot be ruled out, such as a complex, or double stage, irradiation history, any change in primary fluxes like orbital factors, an escape of gaseous products, or an undersaturation of radioactivity. The reputation, however, on radioactive measurements is not high

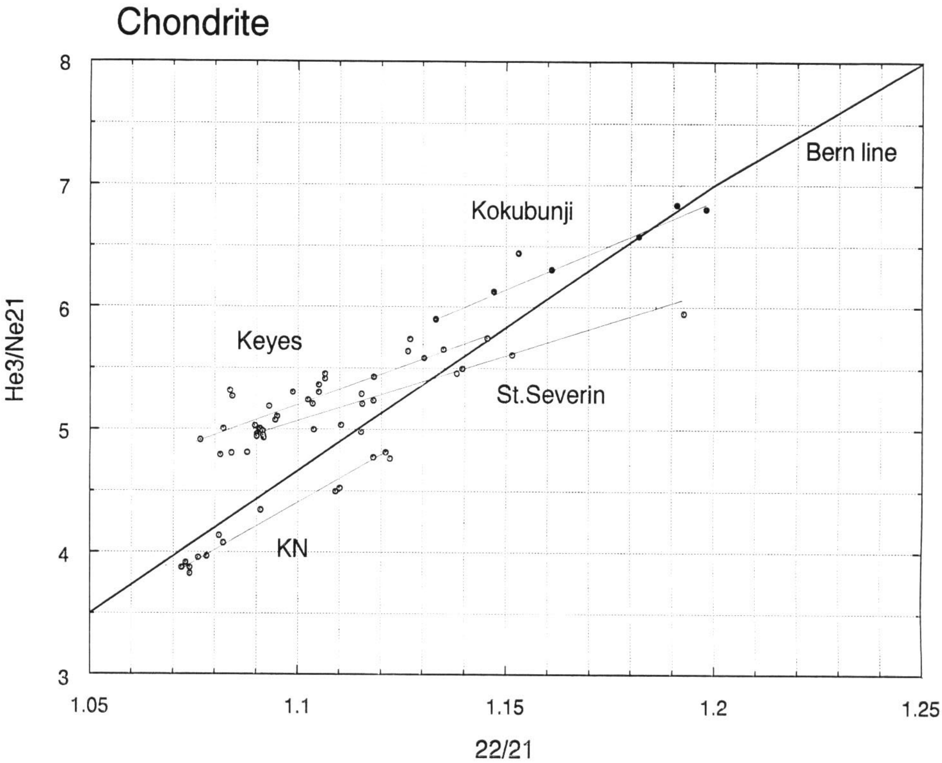

Fig. 3. Shielding of chondrites expressed by noble gases, He-3 and Ne isotopes. (based on Loeken *et al*, 1992).

$^3He/^{21}Ne$ and $^{22}Ne/^{21}Ne$ of chondrites are actually plotted in a two dimensional plane. KN: Knyahinya

$^3He/^{22}Ne = 21.77 - 19.32(^{21}Ne/^{22}Ne)$ for L-chondrites is given by Eugster (1988). In fact, however, the data of depth profiles observed with several chondrites distributed two-dimensionally in a plane of $^3He/^{21}Ne$ and $^{22}Ne/^{21}Ne$. At the center the lowest $^3He/^{21}Ne$ and the lowest $^{22}Ne/^{21}Ne$ are observed.

as those of noble gases; especially for the ratios of high and low energy products overall errors could exceed higher than 10%.

The systematic studies on chondrites can be extended with those of observations of whole rocks of chondrites and of separated metal phases. Chondrites can be treated as a unique target as irons because the compositions are relatively uniform and the corrections are made easily. The extensions from metallic data to those of silicates or chondrites is not difficult, and ^{10}Be, ^{14}C, ^{26}Al, and ^{22}Na can be studied without any difficulty in principle. For these nuclides independent sets of f and $\Delta A'$ must be studied beforehand (Table 1).

Most of high-energy products can be studied only with the metal phase separated from chondrites. The purity of the metal sample must be important. For example,

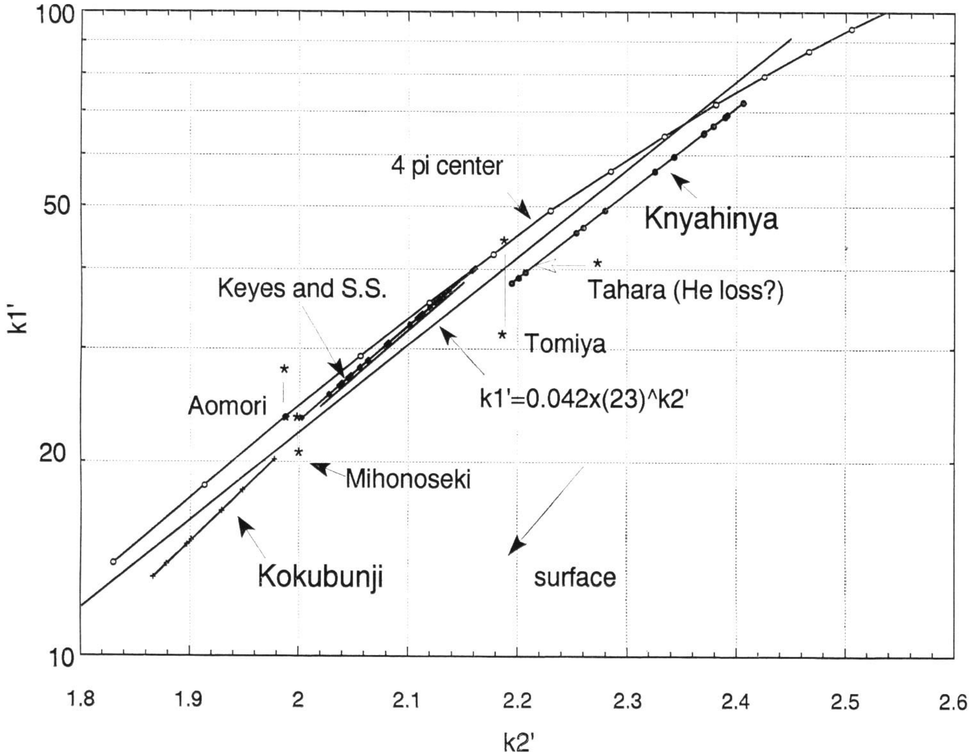

Fig. 4. Shielding of chondrites expressed by k_2' and k_1'.
$k_2' = [\log(22/0.735) - \log(^3He/^{22}Ne)]/\log(23/9.5)$, can be obtained from f and $\Delta A'$ values assigned for ^{3}He and ^{22}Ne respectively. k_2' and k_1' are plotted in a two dimensional plane, but the diagram for smaller meteorites may be approximated by a single line. As described in the text, the production rates of nuclides having $\Delta A' = 23$ are constant throughout whole shielding range.

relative contents of ^{26}Al in chondrite and the metal are usually in the ratios of 20–40, whereas the purifications of the metal can be achieved down to the levels of 0.01–0.1% of stone contaminated in the metal fraction. The level can be examined by determination of typical lithophile elements such as Mg in the solution of the metal fraction after repeated purification steps. In this respect, metal sampling from pallasite is much easier than chondrites and mesosiderites. ^{21}Ne in chondrites and metals are also at the similar ratios, but the metal samples for noble gases cannot be purified at the final step of dissolution in an aqueous solution. This must be the reason why we could not find so far useful data of Ne isotopes in the metal of chondrites. By a similar reason neither ^{4}He nor ^{40}Ar production in chondrite or chondritic metal has been reported. On the other hand, Ar isotopes are usually available in metals, and the ^{38}Ar data, for example, are useful as a substitute of ^{3}He in chondrites. Those of ^{36}Ar are also useful to compare with ^{36}Cl and to calculate exposure age of the chondrites. Only experimental difficulties are in a tedious process to separate and purify these metal phases from chondrite and are in a determination of exact contents of noble gases in these metal samples.

4. Relations between Two Groups of Shielding Indexes.

As described above the explicit expression for shielding can be performed by k_2' and k_1'. On the other hand, for chondrites older expression has been by ^{22}Ne/^{21}Ne and/or ^{3}He/^{21}Ne. The two sets must be compared to unify our understanding for "shielding". We may try to modify first the noble gas data even before to discuss a physical meaning of ^{22}Ne/^{21}Ne. We have two ratis of noble gas data but actually two out of three relatios. ^{3}He/^{22}Ne can be obtained by a slight modification of ^{3}He/^{21}Ne divided by ^{22}Ne/^{21}Ne; the 3/22 is a ratio of two spallation products and k_2' can be obtained, whereas ^{3}He/^{21}Ne is a complex function of the two relations including ^{22}Ne/^{21}Ne. In short, noble gas data can be interpreted as the functions of k_2' and k_1'. There are two alternate choices to relate one of noble gas data to either k_2' or k_1'. First, an attractive idea may be to correlate ^{3}He/^{21}Ne to k_1', because ^{21}Ne/^{3}He is actually proportional to relative $p(^{21}\text{Ne})$, since $p(^{3}\text{He})$ is practically constant under any shielding condition. Then we may correlate 22/21 to k_2' term, which seems not unreasonable, because it is aratio of the two products which is variable with shielding. This first choice, however, may not be a good selection after all, simply because ^{21}Ne/^{3}He increases infinitely with shielding, but actually it is not for k_1'.

The second choice is that 3/21 or the modified figure, may be compared with k_2'. When 3/22 is applied for k_2', composed of logarithmic functions of the ratios of two spallation products, the remaining ^{22}Ne/^{21}Ne or the reciprocal, ^{21}Ne/^{22}Ne, can be plotted against log k_1'. Fig. 5 is an example of the plots of these unified shielding functions. The figure may only mean a superimposition of two figures of Figs. 3 and 4. If so any physical meaning of an apparent correlation of ^{21}Ne/^{22}Ne to log k_1' may not be a serious problem. In fact, however, ^{22}Ne/^{21}Ne can be measured down to 1.05 and no lower data than this barrier are available even under a heavier shielding. Similarly the maximum available k_1', 145 atom/min. g, is also found at the center of a larger body in 4π geometry. It cannot be easily acceptable to correlate k_1' to 22/21, and this barrier may not have any strong background, because we are not quite sure about the physical meaning of 22/21 at this stage.

In fact, as we see in Fig. 5, vertical plots of 21/22 on Y_2 axis are obviously widely distributed than indicated by those of k_1', except for Knyahinya. If the empirical data for 21/22 are correct for the center (and near surface samples) and these different distributions are real, the current method for shielding corrections is not useful enough. This may mean that we have to find some other appropriate correction methods to 21/22 as a function of sample locations in the meteorite before estimating $p(^{21}\text{Ne})$. Or, we have to find an improved relation for systematics for 3/21 and 22/21. The corrected 22/21 or an equation having two variables can provide us with $p(^{21}\text{Ne})$ or

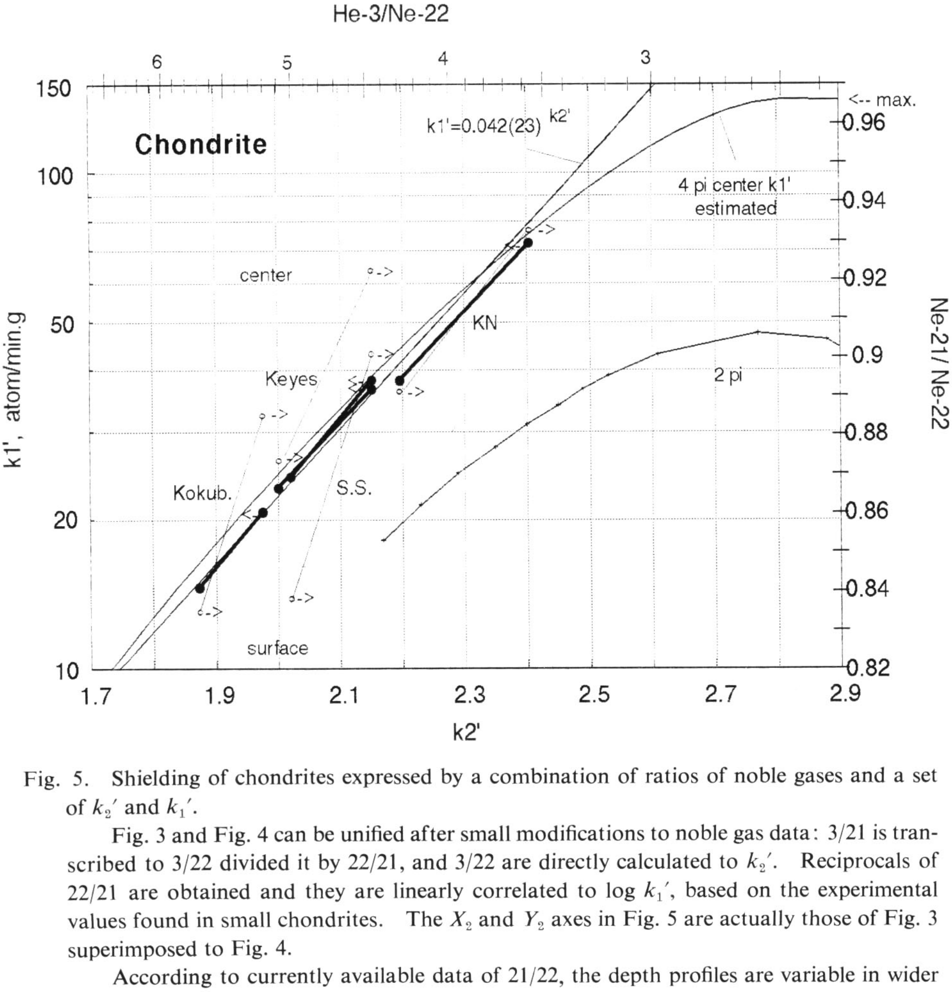

Fig. 5. Shielding of chondrites expressed by a combination of ratios of noble gases and a set of k_2' and k_1'.

Fig. 3 and Fig. 4 can be unified after small modifications to noble gas data: 3/21 is transcribed to 3/22 divided it by 22/21, and 3/22 are directly calculated to k_2'. Reciprocals of 22/21 are obtained and they are linearly correlated to log k_1', based on the experimental values found in small chondrites. The X_2 and Y_2 axes in Fig. 5 are actually those of Fig. 3 superimposed to Fig. 4.

According to currently available data of 21/22, the depth profiles are variable in wider ranges than k_1' estimated and observed directly. This figure therefor displays only an example of fitting of $^{21}Ne/^{22}Ne$. Some more modifications must be made for corrections to 21/22.

other rates. In this situation, the data found in the centers of various size of meteorites must be useful to characterize shielding. Empirically we can normalize 22/21 using some correction factors derived from 3/21 data. This may be nothing but a reduction of 22/21 to estimated values derived from 3/22 at 4π center.

$$21/22 = \log(k_1') \times 0.13 + 0.69\,, \quad \text{and}$$

$$3/22 = 20.96(22/21) - 18.94\,.$$

In other words, only 3/22 can be used as shielding indexes, and 22/21 found at extreme geometries such as near surface must be normalized to reasonable values meeting observed 3/22.

5. Future Studies.

i) The same line of studies will be extended to the productions of nuclides in each target element, such as ^{36}Cl, ^{36}Ar, and ^{38}Ar in Ca, ^{26}Al in Al, ^{7}Be and ^{14}C in oxygen or chondrites. It is always indispensable to have reliable experimental datain a wide range of irradiation conditions.

ii) The lowest energy product involving neutron capture, (n, p), $(n, 2n)$, and (n, α) products may also be studied directly or indirectly in a similar manner.

iii) Quantitative studies on physical meaning of ^{22}Ne/^{21}Ne as the measure of shielding and its limit in the application must be improved. The noble gas data must be collected from the center, or extrapolated to those of center part, as the unique chondritic data for shielding.

It may be interesting to learn that the ^{22}Ne/^{21}Ne has not been reasonably understood yet after 27 years since 1966. This is indeed similar to the case of He isotopes in irons as described in section 2.

iv) These studies can only be extended with reliable experimental data observed in a wide range of meteorites. A limit for the applications of current statistical method cannot be anticipated. Some mixtures of relations for low and high energy products in one or some different targets might give useful results. In some extreme examples we have to abandon any statistical meaning but may still be possible to find useful systematics among the data.

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