Proc. NIPR Symp. Antarct. Meteorites, **5**, 298–309, 1992

NOBLE GASES AND ^{81}Kr-Kr EXPOSURE AGES OF NON-ANTARCTIC ORDINARY CHONDRITES: AN ATTEMPT TO MEASURE TERRESTRIAL AGES OF ANTARCTIC METEORITES

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Abstract: Cosmogenic ^{81}Kr as well as other noble gases of three ordinary chondrites Long Island (L6), Densmore (1950) (H6), and Gladstone (stone) (H6) have been measured with the mass spectrometer newly installed in 1989 to examine the analytical accuracy using the production rates of cosmogenic noble gases proposed by EUGSTER (Geochim. Cosmochim. Acta., **52**, 1649, 1988). For Long Island and Densmore, the ^{81}Kr-Kr ages are 16±2 Ma and 7.7±0.5 Ma, respectively, and are in agreement with the ages by ^{21}Ne and ^{83}Kr, which indicates the validity of both ^{81}Kr analysis and the production rates P_{21} and P_{83} used in this work. Cosmogenic ^{3}He contents in these meteorites are lower than those expected from the production rate P_3, which might be caused by partial loss of He. The ages T_{38} based on ^{38}Ar and P_{38} were shorter than the ages T_{21} and T_{81}. This may be due to the unreasonably high ^{38}Ar production rates calculated for these meteorites, whose low cosmogenic ^{22}Ne/^{21}Ne and ^{78}Kr/^{83}Kr ratios indicate heavy shielding against cosmic-ray irradiation. The constant production rate of cosmogenic ^{38}Ar indicates the better internal concordance than that as a function of shielding depth proposed by EUGSTER. Gladstone is a gas-rich meteorite, for which the abundant solar noble gases made it difficult to estimate the cosmogenic noble gas concentrations and the resulted ages have large uncertainties.

1. Introduction

Cosmogenic ^{81}Kr is an important nuclide for chronological study of meteorites. This radioactive nuclide can be used to determine the cosmic-ray exposure age of meteorites fallen to the earth recently (MARTI, 1967; EUGSTER *et al.*, 1967a; EUGSTER *et al.*, 1987) and the terrestrial age of meteorites of long residence on the earth (SCHULTZ, 1986; FREUNDEL *et al.*, 1986). However, the low concentration of ^{81}Kr in meteorites, $<10^{-13}$ cm^{3}STP/g, makes its accurate measurement difficult.

We are trying to measure ^{81}Kr to determine the terrestrial ages of Antarctic meteorites. Most of the terrestrial ages have been obtained by measuring ^{36}Cl and ^{14}C using AMS (NISHIIZUMI *et al.*, 1989). Cosmogenic ^{81}Kr is also a candidate to determine the terrestrial ages, and has some advantages which are, 1) measurable long terrestrial age of several hundred ky, 2) relatively small sample size less than 1 g, 3) use of noble gas mass spectrometer cheaper than AMS, and 4) the noble gas data provided in addition to ^{81}Kr. Some terrestrial ages based on ^{81}Kr have been reported for Antarctic eucrites and lunar meteorites (*e.g.*, SCHULTZ, 1986; FREUNDEL *et al.*, 1986; Eugster and Niedermann, 1988; Nagao and Ogata, 1989).

Eugster (1988) proposed the production rates of 5 stable cosmogenic nuclides ^{3}He, ^{21}Ne, ^{38}Ar ^{83}Kr, and ^{126}Xe as a function of the shielding dependent ^{22}Ne/^{21}Ne ratio and of the chemical composition. These production rates were determined using the ^{81}Kr-Kr exposure ages and the cosmogenic ^{22}Ne/^{21}Ne ratios measured for 19 ordinary chondrites. In this study, non-Antarctic ordinary chondrites were used to examine the reliability in measuring ^{81}Kr by comparing the cosmic-ray exposure ages calculated by ^{81}Kr and other cosmogenic stable nuclides ^{3}He, ^{21}Ne, ^{38}Ar, ^{83}Kr, and ^{126}Xe using the functions of production rates proposed by Eugster (1988). ^{81}Kr as well as all the noble gas concentrations and isotopic ratios were determined for three chondrites Long Island(L6), Densmore(1950)(H6), and Gladstone(stone) (H6).

2. Samples and Experimental Procedures

The system for noble gas analysis consists of extraction oven, purification line, standard gas system made by Ayumi Industry Co. Ltd. in Japan, and mass spectrometer (VG5400) made by VG Isotopes Limited in England. The noble gas preparation line is evacuated by two turbomolecular pumps to the pressure of 10^{-9} Torr, and the mass spectrometer by an ion pump to 10^{-9} Torr. The mass resolution of the mass spectrometer was adjusted to a $M/\Delta M$ value of about 600.

From 0.6 to 1 g of each sample was wrapped in aluminum foil of 10 μm thick and put into a glass sample holder connected to the ultra-high vacuum extraction and purification line. The line was baked out overnight at the temperatures of 180°C for samples and 250°C for stainless steel extraction and purification line. A known amount of atmosphere was analyzed with the same procedure as applied to the meteorite sample to determine the sensitivity and the mass discrimination for each noble gas element. He standard gas, of which ^{3}He/^{4}He ratio was 1.71×10^{-4}, was used for the determination of mass discrimination for He. Amounts of noble gases in blank run were 6×10^{-9}, 5×10^{-11}, 1×10^{-11}, 5×10^{-13}, and 1×10^{-13} cm^{3}STP for ^{4}He, ^{20}Ne, ^{36}Ar, ^{84}Kr, and ^{132}Xe, respectively.

The sample to be measured was dropped into molybdenum crucible in the extraction oven and melted by heating at 1700°C for 15 min. The extracted noble gases were purified by removing reactive gases such as hydrocarbon, oxygen, and so on by Ti-Zr getter heated at about 800°C, and the purified noble gases were separated into three fractions He-Ne, Ar, and Kr-Xe, for mass spectrometry using charcoal trap controlled at the temperatures of liquid air and -60°C. Hydrocarbon peak of mass number 81 was separated from the peak of ^{81}Kr. Since ^{79}Br peak was lower than the detection limit, the interference of ^{81}Br peak at ^{81}Kr could be negligible in the Kr analyses. These results suggested that the detection limit of ^{81}Kr was about 1×10^{-14} cm^{3}STP under the present condition of this mass spectrometer.

3. Results and Discussion

Noble gas analysis was done four and two times for Long Island(L6) and Densmore(1950)(H6), respectively. A single analysis was performed for Gladstone (stone)(H6). All noble gas data for He, Ne, and Ar are listed in Table 1, in which the data of Long Island by HINTENBERGER *et al.* (1964) and ZÄHRINGER (1966) are included for comparison. The He, Ne, and Ar isotopic compositions and concentrations measured in this work are in agreement with those by them. For Kr and Xe, only the representative concentrations and isotopic ratios are presented in Table 2. ^{81}Kr could be measured for these meteorite. The ^{81}Kr concentrations were 2.6, 1.9, and 2.1×10^{-14} cm^{3}STP/g in Long Island, Densmore, and Gladstone, respectively. Statistical errors for the isotopic ratios are 1σ and the uncertainties for the concentrations are estimated at about 10%. Ne isotopic ratios of Long Island and Densmore imply that Ne in these meteorites is totally cosmogenic. The high ^{3}He/^{4}He ratios also suggest the pure cosmogenic product for ^{3}He in these meteorites. Contrary to these meteorites, Gladstone has large amounts of light noble gases, He, Ne, and Ar, of which the elemental compositions and Ne isotopic ratios strongly suggest the solar wind origin for these noble gases.

3.1. ^{81}Kr-Kr exposure ages

Cosmic-ray exposure age based on ^{81}Kr is calculated by the equation (MARTI, 1967; EUGSTER *et al.*, 1967a),

$$T_{81}=(1/\lambda)(P_{81}/P_{83})(^{83}\mathrm{Kr}/^{81}\mathrm{Kr})_c,$$

where $\lambda\,(=3.25\times10^{-6}\,\mathrm{y}^{-1})$ is the decay constant of ^{81}Kr (EASTWOOD *et al.*, 1964), P_{81} and P_{83} are the production rates of cosmogenic ^{81}Kr and ^{83}Kr, respectively. The production rate ratio P_{81}/P_{83} depends on the composition of target elements and on the shielding depth, and it can be estimated by the following equations (MARTI, 1967; MARTI and LUGMAIR, 1971; FINKEL *et al.*, 1978; EUGSTER, 1988);

1) $P_{81}/P_{83}=(0.95/2)(^{80}\mathrm{Kr}/^{83}\mathrm{Kr}+{}^{82}\mathrm{Kr}/^{83}\mathrm{Kr})_c$,
2) $P_{81}/P_{83}=1.262(^{78}\mathrm{Kr}/^{83}\mathrm{Kr})_c+0.381$, and
3) $P_{81}/P_{83}=0.562(^{22}\mathrm{Ne}/^{21}\mathrm{Ne})_c-0.029$.

The third equation was proposed for ordinary chondrites (EUGSTER, 1988). The isotopic ratios of cosmogenic Kr and the production rate ratios P_{81}/P_{83} obtained for Long Island and Densmore using the above three equations are summarized in Table 3. However, for gas-rich meteorite Gladstone, the ratio could not be determined. The isotopic ratios of cosmogenic Kr were calculated by subtracting AVCC-Kr from measured Kr assuming no fissiogenic Kr components in these meteorites. The P_{81}/P_{83} ratios calculated by the first equation were obviously higher than those calculated by other two equations because of a production of ^{40}Ar ions interfering ^{80}Kr by charge transfer reaction in the flight tube of mass spectrometer. Possibility of neutron capture by ^{79}Br for the high ^{80}Kr can be eliminated since the ^{82}Kr/^{84}Kr ratios of these meteorites show no obvious excess due to the neutron capture by ^{81}Br. Therefore, the calculated ratio using the first equation must be an overestimation owing to the high ^{80}Kr/^{83}Kr ratio. On the other hand, systematic differences between

Table 1. Concentrations and isotopic ratios of He, Ne, and Ar in three ordinary chondrites.

Sample	weight(g)	^{3}He	^{4}He	^{3}He/^{4}He	^{20}Ne	^{21}Ne	^{22}Ne	^{20}Ne/^{22}Ne	^{21}Ne/^{22}Ne	^{36}Ar	^{38}Ar	^{40}Ar	^{38}Ar/^{36}Ar	^{40}Ar/^{36}Ar
Long Island	1.01	152	3640	0.0417	54.0	59.7	62.9	0.8588	0.9436	12.5	7.03	19100	0.5494	1523
(L6)				±.0001				±.0118	±.0086				±.0056	±5
	1.19	154	3610	0.0426	54.9	61.1	64.4	0.8527	0.9488	13.0	7.04	19500	0.5419	1498
				±.0001				±.0054	±.0025				±.0055	±6
	0.825	196	4750	0.0412	74.4	83.3	88.2	0.8431	0.9446	16.1	8.27	23300	0.5150	1451
				±.0017				±.0007	±.0014				±.0032	±8
	0.657	166	4700	0.0353	50.0	56.1	59.9	0.8349	0.9372	12.9	6.56	18200	0.5089	1411
				±.0005				±.0039	±.0085				±.0057	±16
(Reference)		205	3700		62.1	54.3	56.5							1)
		210	4700		45.7	45.0	48.0			13.3	6.1	15400		2)
Densmore (1950)	1.01	62.6	931	0.0672	23.1	25.4	27.0	0.8562	0.9412	5.75	3.21	1840	0.5584	320.1
(H6)				±.0002				±.0115	±.0238				±.0053	±1.0
	0.586	73.7	1350	0.0546	28.9	31.8	34.1	0.8477	0.9313	5.47	2.94	1890	0.5367	345.8
				±.0007				±.0040	±.0111				±.0043	±1.4
Gladstone (stone)	1.12	173	210000	0.000827	5690	54.9	521	10.92	0.1053	335.4	71.4	45800	0.2129	136.56
(H6)				±.000008				±.03	±.0023				±.0020	±.35

Concentrations of He, Ne and Ar are given in the unit of 10^{-9}cm^{3}STP/g.

1) HINTENBERGER *et al.* (1964) and 2) ZÄHRINGER (1966) in compilation by SCHULTZ and KRUSE (1989).

Table 2. Concentrations and isotopic ratios of Kr and Xe in three ordinary chondrites.

Sample	^{84}Kr*	^{78}Kr	^{80}Kr	^{81}Kr	^{82}Kr	^{83}Kr	^{86}Kr	^{132}Xe*	^{124}Xe	^{126}Xe	^{128}Xe	^{129}Xe	^{130}Xe	^{131}Xe	^{134}Xe	^{136}Xe
		^{84}Kr=100							^{132}Xe=100							
Long Island	85.5	0.928	5.21	0.030	21.70	22.36	30.23	45.6	0.547	0.564	9.29	108.5	16.13	81.97	38.42	32.25
(L6)		±.026	±.04	±.010	±.18	±.19	±.11		±.017	±.013	±.05	±.4	±.09	±.37	±.14	±.16
Densmore (1950)	83.7	0.758	4.42	0.023	20.91	21.00	30.55	40.8	0.501	0.477	8.07	110.4	15.94	81.11	38.61	32.43
(H6)		±.020	±.06	±.007	±.09	±.12	±.22		±.021	±.022	±.08	±.4	±.09	±.29	±.15	±.12
Gladstone (stone)	425	0.700	5.33	0.005	20.91	20.81	30.81	383	0.457	0.438	8.32	122.2	16.25	81.56	38.30	32.02
(H6)		±.009	±.05	±.001	±.08	±.07	±.13		±.008	±.022	±.04	±.3	±.05	±.13	±.20	±.17
AVCC		0.597	3.02		20.15	20.17	30.98$^{1)}$		0.464	0.416	8.21	104.5	16.23	82.06	38.08	31.98$^{2)}$

*Concentrations of ^{84}Kr and ^{132}Xe are given in the unit of 10^{-12} cm^{3}STP/g. Reference: 1) EUGSTER *et al.* (1967b). 2) PODOSEK *et al.* (1971).

Table 3. Isotopic ratios of cosmogenic Kr, calculated production rate ratios and ^{81}Kr-Kr exposure ages.

Sample	Weight	^{78}Kr	^{80}Kr	^{81}Kr	^{82}Kr	^{84}Kr	P_{81}/P_{83}*			T_{81} (Ma)**	
	(g)	^{83}Kr=100					1)	2)	3)	2)	3)
Long Island	1.01	15.6	97.9	1.03	72.3	86.0	0.808	0.578	0.563	17.3	16.8
(L6)		±.6	±3.1	±.41	±4.7	±15.4	±.027	±.008	±.006	±6.0	±6.7
	1.19	12.5	63.1	1.07	74.3	87.3	0.653	0.539	0.563	15.5	16.2
		±.5	±2.6	±.16	±3.5	± 8.7	±.021	±.006	±.002	±2.3	±2.4
	0.825	12.1	48.9	1.00	78.1	87.6	0.603	0.534	0.566	16.4	17.4
		±3.6	±2.6	±.22	±4.9	±12.4	±.026	±.045	±.001	±3.8	±3.8
	0.657	12.9	53.2	1.17	78.5	93.4	0.626	0.543	0.571	14.3	15.0
		±1.4	±4.6	±.41	±9.9	±15.9	±.052	±.018	±.006	±5.0	±5.3
										mean 16	
										±2 (2σ)	
Densmore	1.01	13.9	98.0	2.31	82.7	119.6	0.858	0.557	0.568	7.43	7.56
(1950)		±1.2	±8.1	±.31	±14.5	±25.1	±.079	±.015	±.015	±1.28	±1.03
(H6)	0.586	15.3	54.8	2.22	101.9	136.8	0.744	0.574	0.575	7.95	7.96
		±3.5	±11.9	±.78	±25.5	±74.2	±.159	±.044	±.007	±2.84	±2.80
										mean 7.7	
										±.5 (2σ)	

* Production rate ratios were calculated using the equations 1), 2), and 3) presented in the text.
** Cosmic-ray exposure ages 2) and 3) were obtained using the production rate ratios 2) and 3), respectively.

the production rate ratios calculated using the second and third equations could not be noticed. For this reason, only the cosmic-ray exposure ages calculated using the production rate ratios estimated from the latter two equations were presented in Table 3. For each sample, the exposure ages were in good agreement with each other within experimental errors. The mean values are 16±2 Ma for Long Island and 7.7±0.5 Ma for Densmore. The statistical errors are 2σ.

3.2. Cosmic-ray exposure ages based on stable noble gases

Concentrations of cosmogenic noble gases, cosmogenic ^{22}Ne/^{21}Ne ratios, production rates, and cosmic-ray exposure ages are summarized in Table 4. All ^{3}He and ^{21}Ne of Long Island and Densmore were assumed to be the spallogenic products by cosmic-ray irradiation. For Gladstone, the cosmogenic ^{3}He content was calculated assuming the observed ^{4}He to be of solar wind origin with the ^{3}He/^{4}He ratio of 4×10^{-4}. Cosmogenic ^{21}Ne content of this meteorite was calculated using a two-component mixing model between solar and cosmogenic Ne, in which ^{21}Ne/^{22}Ne ratios of 0.031 (EBERHARDT *et al.*, 1972) and 0.918 (MAZOR *et al.*, 1970) were assumed for solar and cosmogenic Ne, respectively. In the calculation of cosmogenic ^{38}Ar, ^{83}Kr, and ^{126}Xe concentrations, the following isotopic ratios were adopted (EUGSTER *et al.*, 1967b; PODOSEK *et al.*, 1971): $({}^{38}\mathrm{Ar}/{}^{36}\mathrm{Ar})_c=1.6$, $({}^{38}\mathrm{Ar}/{}^{36}\mathrm{Ar})_t=0.188$, $({}^{83}\mathrm{Kr}/{}^{86}\mathrm{Kr})_t=0.6511$, $({}^{126}\mathrm{Xe}/{}^{132}\mathrm{Xe})_c=2$, $({}^{126}\mathrm{Xe}/{}^{132}\mathrm{Xe})_t=0.00416$, $({}^{136}\mathrm{Xe}/{}^{132}\mathrm{Xe})_t=0.3198$, and $({}^{136}\mathrm{Xe}/{}^{132}\mathrm{Xe})_f=1.130$. For Gladstone, the concentrations of cosmogenic ^{83}Kr and ^{126}Xe could not be estimated owing to the high concentrations

Table 4. Concentrations of cosmogenic noble gases, cosmogenic ^{22}Ne/^{21}Ne ratios, production rates, and cosmic-ray exposure ages.

Sample	$(^3He)_c$	$(^{21}Ne)_c$	$(^{38}Ar)_c$	$(^{83}Kr)_c$	$(^{126}Xe)_c$	$\left(\frac{^{22}Ne}{^{21}Ne}\right)_c$	P_3	P_{21}	P_{38}	(P_{38})*
	$\times 10^{-9}$cm^{3}STP/g			$\times 10^{-12}$cm^{3}STP/g			$\times 10^{-9}$cm^{3}STP/g·Ma			
Long Island	167	65	5.27	2.22	0.0675	1.058	16.4	4.34	0.50	0.40
(L6)	±20	±12	±.53	±.16	±.0059	±.006				
Densmore (1950)	68	29	2.30	0.86	0.0249	1.068	16.3	4.10	0.53	0.43
(H6)	±8	±5	±.18	±.10	±.0082	±.008				
Gladstone (stone)	84	40	9.50				16**	3.1**	0.50	0.43
(H6)	±9	±8	±.95							

Sample	P_{83}	P_{126}	T_3	T_{21}	T_{38}	(T_{38})*	T_{83}	T_{126}	T_{81}
	$\times 10^{-12}$cm^{3}STP/g·Ma		Ma						Ma
Long Island	0.156	0.00746	10	15	11	13	14	9	16
(L6)			±1	±3	±1	±1	±1	±2	±2
Densmore (1950)	0.148	0.00736	4.2	7.0	4.3	5.1	5.8	3.4	7.7
(H6)			±.5	±1.1	±.4	±.4	±.7	±1.1	±.5
Gladstone (stone)			5.3	13	22				
(H16)			±.6	±3	±2				

Cosmogenic noble gas concentrations of He, Ne and Ar for Long Island and Densmore (1950) are mean values.

Production rates are calculated using following equations proposed by EUGSTER (1988) (in the unit of $\times 10^{-8}$cm^{3}STP/g·Ma): $P_3=F[2.09-0.43\ (^{22}Ne/^{21}Ne)_c]$, $(F_L=1.00, F_H=0.98)$; $P_{21}=1.61F[21.77(^{22}Ne/^{21}Ne)_c-19.32]^{-1}$, $(F_L=1.00,\ F_H=0.93)$; $P_{38}=F[0.125-0.071(^{22}Ne/^{21}Ne)_c]$, $(F_L=1.00, F_H=1.08)$; $P_{83}=0.0196F[0.62\ (^{22}Ne/^{21}Ne)_c-0.53]^{-1}$, $(F_L=1.00,\ F_H=1.00)$; $P_{126}=F[0.0174-0.0094\ (^{22}Ne/^{21}Ne)_c]$, $(F_L=1.00,\ F_H=1.00)$.

* $P_{38}=0.4F\times 10^{-9}$cm^{3}STP/g·Ma $(F_L=1.00,\ F_H=1.08)$ is used (see text).

** Production rates for Gladstone (stone) are calculated with assumption that $(^{22}Ne/^{21}Ne)_c=1.11$.

of trapped Kr and Xe. The average values of cosmogenic noble gas concentrations are presented in Table 4.

Cosmic-ray exposure ages based on the cosmogenic ^{3}He, ^{21}Ne, ^{38}Ar, ^{83}Kr, and ^{126}Xe were calculated using the production rates by EUGSTER (1988). The formulas are given in the caption of Table 4. However, the production rates P_{38} by EUGSTER (1988) seem to be too high as discussed later.

The ^{22}Ne/^{21}Ne ratios of Long Island and Densmore are 1.058±0.006 and 1.068±0.008, respectively (Table 4). The cosmogenic ^{78}Kr/^{83}Kr ratios were also as low as 0.13 and 0.14 for Long Island and Densmore, respectively (in Table 3). These low ratios are plotted on the correlation line between the ^{22}Ne/^{21}Ne and ^{78}Kr/^{83}Kr ratios for chondrites presented by EUGSTER (1988), strongly implying the heavy shielding against cosmic-rays for these meteorites. The recovered mass of Long Island was more than 600 kg (GRAHAM *et al.*, 1985), which corresponds to a preatmospheric body larger than 70 cm in diameter. Whereas, the recovered mass of Densmore was not so large.

The exposure ages T_{21} by ^{21}Ne are 15±3 and 7.0±1.1 Ma for Long Island and Densmore, respectively, which are in good agreement with the ^{81}Kr-Kr ages T_{81} as shown in Table 4. This confirms the ^{21}Ne production rate by EUGSTER (1988).

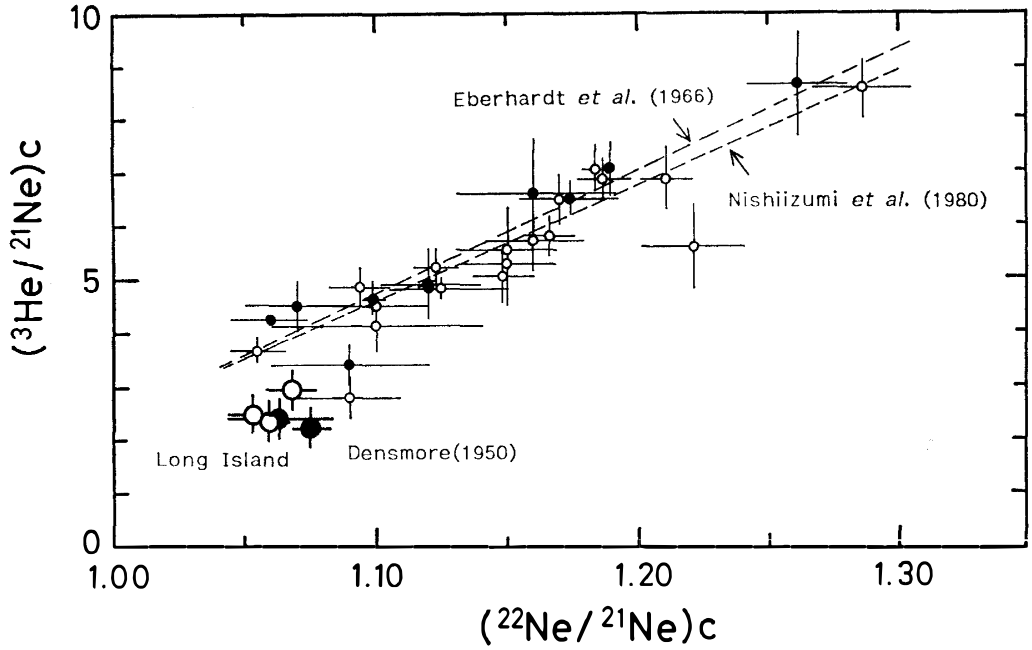

Fig. 1. Correlation plot between cosmogenic $^{3}He/^{21}Ne$ and $^{22}Ne/^{21}Ne$ ratios of ordinary chondrites reported by EUGSTER *(1988). The correlation lines plotted were proposed by* EBERHARDT *et al. (1966) and* NISHIIZUMI *et al. (1980). Open symbol and solid symbol correspond to L- and H- chondrites, respectively. Long Island and Densmore are plotted below the line, suggesting ^{3}He loss from these meteorites. The low $^{22}Ne/^{21}Ne$ ratios of Long Island and Densmore indicate the heavy shielding against cosmic-ray irradiation.*

However, the exposure ages T_3 by ^{3}He are shorter than those by ^{21}Ne. Because the ^{3}He/^{21}Ne and ^{22}Ne/^{21}Ne ratios for these meteorites are plotted below the correlation line for ordinary chondrites as shown in Fig. 1, these short ages might be caused by the partial loss of ^{3}He from these meteorites.

The T_{83} age of Long Island agrees with the age T_{81} within the experimental errors. While, T_{83} of Densmore is somewhat younger than T_{81}. Although the ages T_{126} of Long Island and Densmore are about a half of the ages T_{21} and T_{81}, we could not find out the reason why the T_{126} ages are so young in comparison with the other ages. For Gladstone, concordant exposure ages T_3, T_{21} and T_{38} could not be obtained.

3.3. ^{38}Ar production rate

Because the ^{38}Ar production rates calculated using the formula proposed by EUGSTER (1988) were as high as 0.50 and 0.53×10^{-9} cm^{3}STP/gMa for Long Island and Densmore, the ages T_{38} calculated using these production rates were 10.5 and 4.3 Ma, respectively, which were 30 and 45% shorter than the T_{81} ages. SCHULTZ *et al.* (1991) proposed the lower value of production rate than that by EUGSTER (1988) for H-chondrites. However, the exposure ages calculated for Long Island and Densmore using the reduced production rate were still 20 and 30% shorter than the ages T_{81} and T_{21}, respectively. Since the He, Ne, and Ar isotopic ratios and concentrations of Long Island are practically the same as those previously

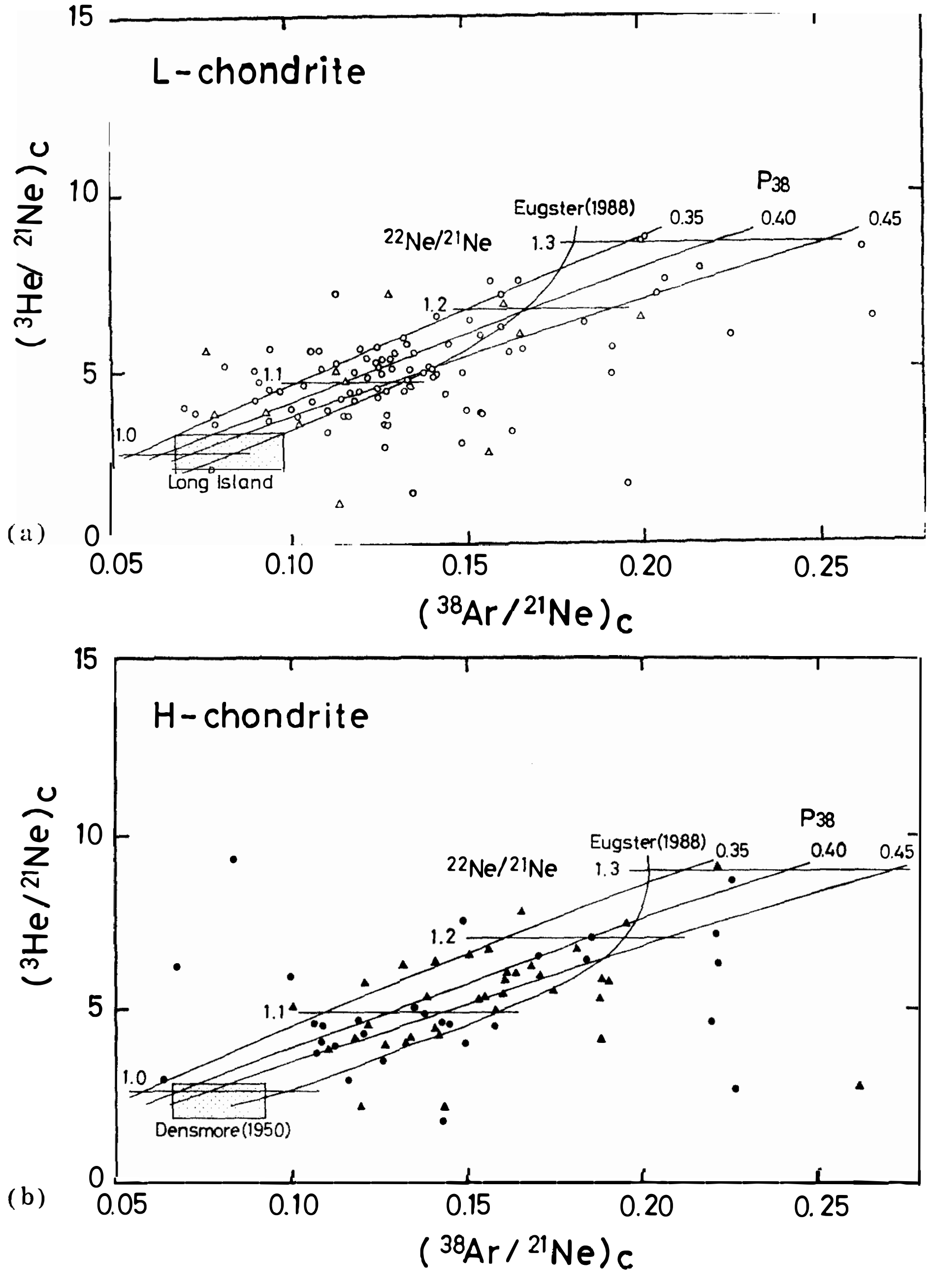

Fig. 2a, b. *Plots of cosmogenic ^{3}He/^{21}Ne ratio versus ^{38}Ar/^{21}Ne ratio for L- and H-chondrites. Most of data compiled by Schultz and Kruse (1989), Eugster (1988), and Schultz et al. (1991) are plotted. Circle and triangle symbols show the data from non-Antarctic and Antarctic chondrites, respectively. Differences between the distribution patterns of Antarctic and non-Antarctic chondrites could not be found. The data points show a positive correlation, which has resulted from the various shielding depths. Since Long Island and Densmore (1950) are plotted on the left lower side with low ^{3}He/^{21}Ne and ^{38}Ar/^{21}Ne ratios, the heavy shielding is suggested for these meteorites. It is supported by the large recovered mass, >600 kg, for Long Island. The curved lines labeled as Eugster were calculated using the formulas to calculate production rates P_3, P_{21}, and P_{38} by Eugster (1988), and numerical figures along the lines represent the ^{22}Ne/^{21}Ne ratios. Other lines were calculated assuming the constant ^{38}Ar production within meteorite, where P_{38} values of 0.35, 0.40, and 0.45 $\times 10^{-9}$cm^{3}STP/gMa were used. These lines fit well compared with the curved line based on the production rates by Eugster (1988) (see text).*

reported by HINTENBERGER *et al.* (1964) and ZÄHRINGER (1966) (in Table 1), the short exposure ages based on ^{38}Ar are difficult to be attributed to underestimation of ^{38}Ar concentrations in the Ar analysis in this study. As already described, the cosmogenic Ne and Kr isotopic ratios imply the heavy shielding against cosmic-rays for these meteorites. The short T_{38} ages seem to have resulted from the high ^{38}Ar production rates calculated using the low ^{22}Ne/^{21}Ne ratios of about 1.06.

The formula P_{38} as a function of ^{22}Ne/^{21}Ne ratio was deduced from the correlation plot between P_{38} and ^{22}Ne/^{21}Ne ratio (Fig. 7 in EUGSTER, 1988). However, the negative correlation as expressed by the formula is very vague. The nearly constant concentrations of cosmogenic ^{38}Ar within Keyes (WRIGHT *et al.*, 1973) and St. Severin (SCHULTZ and SIGNER, 1976) suggest the negligible shielding effect on the ^{38}Ar production even in a meteorite of 50 cm in diameter.

Moreover, the production rates P_3, P_{21}, and P_{38} as a function of ^{22}Ne/^{21}Ne ratio by EUGSTER (1988) are not concordant internally. Figures 2a and b are the plots of cosmogenic ^{3}He/^{21}Ne ratio *versus* ^{38}Ar/^{21}Ne ratio for L- and H-chondrites of petrologic types 5 and 6. Most of the ratios in the compilation by SCHULTZ and KRUSE (1989), and the data by EUGSTER (1988) and SCHULTZ *et al.* (1991) are plotted. The data points show a positive correlation, and the ratios ^{3}He/^{21}Ne and ^{38}Ar/^{21}Ne are in the ranges of 1–9 and 0.06–0.27, respectively. The curved lines labeled as EUGSTER were calculated using the production rates by EUGSTER (1988), and the numerical figures along the lines represent the ^{22}Ne/^{21}Ne ratio. If we consider the cosmogenic ^{22}Ne/^{21}Ne ratio from 1.06 to 1.30, the calculated ^{38}Ar/^{21}Ne ratio varies from 0.115 to 0.18 and from 0.135 to 0.20 for L and H chondrites, respectively, which cannot cover the range of ^{38}Ar/^{21}Ne ratios already reported for ordinary chondrites. An unusually low cosmogenic ^{22}Ne/^{21}Ne ratio of less than 1.05 is demanded for the data plotted in the lower left area corresponding to the heavy shielding. This discordance seems to arise from the negative correlation line between P_{38} and ^{22}Ne/^{21}Ne ratio (EUGSTER, 1988) which increases P_{38}/P_{21} with decrease in ^{22}Ne/^{21}Ne ratio. The upper limit for P_{38}/P_{21} ratio has also resulted from the negative correlation.

We approximated the ^{38}Ar production rate as constant within the meteorites and calculated the correlation lines between P_3/P_{21} and P_{38}/P_{21} ratios assuming the P_{38} as from 0.35 to 0.45×10^{-9} cm^{3}STP/gMa, which are presented in Figs. 2a and b. The lines fit well to the data field of L and H chondrites, and the P_{38}/P_{21} ratio corresponding to ^{22}Ne/^{21}Ne ratio from 1.06 to 1.30 spans the range for most of the data points. With this production rate, the internal concordance has been largely improved, which indicates that the constant production of cosmogenic ^{38}Ar within meteorites is better approximation than the function of the shielding dependent ^{22}Ne/^{21}Ne ratio.

We tentatively adopted the production rates P_{38} of 0.40 and 0.43×10^{-9} cm^3 STP/gMa for L and H chondrites, respectively. The cosmic-ray exposure ages calculated for Long Island, Densmore, and Gladstone are 13±1, 5.1±0.4, and 22±2 Ma, respectively, which are still shorter than the ages T_{21} and T_{81}. These discordances might be caused by unusually heavy shielding depth where the production of ^{38}Ar decreases.

3.4. Can we measure the terrestrial ages of Antarctic meteorites by ^{81}Kr method with the mass spectrometer?

As already described the ^{81}Kr-Kr age of meteorite recently fallen to the earth means the cosmic-ray exposure age of this meteorite, the age of which should be in agreement with the exposure ages derived from other method using the stable cosmogenic nuclides. The object of this study was to examine the reliability in analysis of a very small amount of ^{81}Kr with the mass spectrometer installed in our laboratory.

For Long Island and Densmore, the ages T_{21} agree with the ages T_{81}. Since the production rate of ^{21}Ne in ordinary chondrites has been determined by different radioactive nuclide and radioactive-stable pairs such as ^{53}Mn, ^{81}Kr-^{83}Kr, and ^{22}Na-^{22}Ne, and the rate has been confirmed by several authors (*e.g.*, NISHIZUMI *et al.*, 1980; MÜLLER *et al.*, 1981), P_{21} used in this work may not be largely changed. If we accept the exposure gae based on cosmogenic ^{21}Ne as the true age, the agreement between the ages T_{21} and T_{81} supports the validity in measurement of small quantity of ^{81}Kr, 10^{-14} cm^{3}STP, in meteorites using our mass spectrometer.

Detection limit of ^{81}Kr with this mass spectrometer was about 1×10^{-14} cm^{3}STP. In this experimental condition, ^{81}Kr of some meteorites such as eucrites with abundant target elements is measurable. However, if this method is applied to the Antarctic ordinary chondrites with the terrestrial ages older than the half-life of ^{81}Kr (2.1×10^5 y), the amount of ^{81}Kr extracted from a gram-size of specimen will be the same as the detection limit or less. Thus, the application of the ^{81}Kr method to Antarctic ordinary chondrites for the determination of their terrestrial ages demands the improved detection limit for the mass spectrometer in our laboratory.

4. Summary

1) We could detect ^{81}Kr for three ordinary chondrites Long Island(L6), Densmore(1956)(H6), and Gladstone(stone)(H6) by the mass spectrometer recently installed in our laboratory. The concentrations were about 2×10^{-14} cm^{3}STP/g, which was two times the detection limit of this mass spectrometer. The cosmic-ray exposure ages by the ^{81}Kr-Kr method were calculated as 16±2 and 7.7±0.5 Ma for Long Island and Densmore, respectively, and were compared with the exposure ages calculated by ^{3}He, ^{21}Ne, ^{38}Ar, ^{83}Kr, and ^{126}Xe. The ages T_{81} are in consistent with the ages T_{21}, indicating the accuracy of ^{81}Kr analysis.

2) Since Gladstone(stone) is a gas-rich meteorite, cosmogenic noble gases were masked with the large amounts of trapped solar type noble gases, for which the precise estimation of abundances of cosmogenic noble gas isotopes was difficult. The age T_{81} could not be obtained and the exposure ages T_3, T_{21}, and T_{38} did not agree with each other.

3) In the plot of ^{3}He/^{21}Ne ratio versus ^{38}Ar/^{21}Ne ratio for ordinary chondrites, positive correlation depending on the shielding effect was found. The formula P_{38} as a function of cosmogenic ^{22}Ne/^{21}Ne ratio proposed by EUGSTER (1988) cannot explain the field where the data are plotted. If we assume the constant ^{38}Ar production within meteorite, the calculated correlation lines fit well to the distribution

of data points. The exposure ages T_{38} calculated assuming the constant ^{38}Ar production rate show better agreement with T_{21} and T_{81} than those obtained with the formula by EUGSTER (1988).

4) Since ^{81}Kr of Antarctic meteorites with abundant target elements is measurable with our mass spectrometer, the terrestrial ages can be determined for these meteorites by ^{81}Kr-Kr method. However, an improvement in detection limit is needed for the terrestrial ages of Antarctic ordinary chondrites.

Acknowledgments

The authors would like to thank Miss A. OGATA of the Institute for Study of the Earth's Interior, Okayama University for her assistance in sample preparation and noble gas analysis. Thanks are also expressed to Prof. S. AKIMOTO of the Institute for Study of the Earth's Interior, Okayama University for his advice and encouragement for our study.

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(*Received August 5, 1991; Revised manuscript received October 11, 1991*)