und gravimetrischen [24]:

$$D_H = 1{,}0 \cdot 10^{-6}\ \text{cm}^2\ \text{s}^{-1}$$

Messungen mit leichtem Wasserstoff. Sowohl die Messungen mit Tritium als auch die röntgenographischen und gravimetrischen Methoden sind von Durchtrittshemmungen unabhängig. Die Übereinstimmung der Ergebnisse macht deutlich, daß früher für 25 °C veröffentlichte Werte von 10^{-9} oder gar 10^{-20} cm^2 s^{-1} auf den Einfluß von Grenzflächenreaktionen zurückzuführen sind.

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Herrn Prof. Dr. E. WICKE möchten wir an dieser Stelle für wertvolle Hinweise und anregende Diskussionen herzlich danken. — Der Deutschen Forschungsgemeinschaft danken wir für die Unterstützung unserer Arbeit mit Personal- und Sachmitteln.

Mass Fractionation and the Isotopic Anomalies of Xenon and Krypton in Ordinary Chondrites

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(Z. Naturforsch. **26 a**, 1980—1986 [1971]; received 7 August 1971)

The abundance and isotopic composition of all noble gases are reported in the Wellman chondrite, and the abundance and isotopic composition of xenon and krypton are reported in the gases released by stepwise heating of the Tell and Scurry chondrites. Major changes in the isotopic composition of Xe result from radiogenic Xe^{129} and from variations in the isotopic mass fractionation pattern in the different temperature fractions. The isotopic composition of trapped krypton in the different temperature fractions of the Tell and Scurry chondrites displays smaller fractional changes than xenon, but the isotopic composition of these two gases covary in the manner expected from mass dependent fractionation.

Many clues of early geologic events have been recorded in the stable noble gases, He, Ne, Ar, Kr and Xe. The chemical inertness and volatile nature, even at relatively low temperatures, resulted in almost complete loss of these elements from more condensable material when solid planetary matter formed in our solar system. Many isotopic anomalies of these elements have been generated in solid planetary material subsequent to this separation, by induced nuclear reactions and by natural radioactive decay of more abundant elements.

Although the generation of isotopic anomalies of all the stable noble gases by nuclear processes is well documented, there is also evidence for the production of isotopic anomalies of noble gases by simple isotopic mass fractionation at the time of the separation of these from more condensable elements. In 1949 SUESS [1] and BROWN [2] independently noted that a comparison of the earth's inventory of noble gases with the solar abundances of these elements shows preferential loss from terrestrial material of the gases lighter than krypton. Subsequent work on noble gases in terrestrial sediments [3] found high concentrations of xenon which, when added to the atmospheric inventory of noble gases, show that the earth's fractionation of noble gases extends to the heaviest stable gas, xenon. The trapped noble gases in stone meteorites display a similar fractionation pattern [4, 5] with the xenon preferentially retained over neon by a factor $\approx 10^4 - 10^5$.

SIGNER and SUESS [6] discussed the role of mass fractionation in generating isotopic anomalies of neon and argon in planetary material. They noted a covariance of Ar^{36}/Ar^{38}, Ne^{20}/Ne^{22} and Ne^{20}/Ar^{36} ratios in atmospheric and meteoritic gases. MANUEL [7] reported variations in the trapped Ne^{20}/Ne^{22}

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and Ne^{21}/Ne^{22} ratios in the different temperature fractions of the Fayetteville meteorite and suggested that these variations result from isotopic mass fractionation. However, PEPIN and coworkers [8–10] reported that there is no sign that diffusive fractionation is responsible for any of the isotopic variations of meteoritic neon. Recent results on the isotopic composition of trapped meteoritic He, Ne and Ar in the laboratories at Minnesota, Chicago and Rice have been attributed to nuclear reactions [10–14], but other authors [15–17] have maintained that the observed variations result from simple isotopic fractionation.

For the heavy noble gases, a similar divergence of opinions has developed. SRINIVASAN et al. [18] first suggested that spontaneous fission of superheavy elements may be responsible for the enrichment of heavy xenon isotopes in carbonaceous chondrites. ANDERS and HEYMANN [19] have arrived at a similar conclusion by comparing the excess heavy xenon isotopes with the content of volatile elements in carbonaceous chondrites, and EBERHARDT et al. [20] have interpreted the isotopic composition of solar-type xenon as evidence for large amounts of fissiogenic xenon in carbonaceous chondrites. However, KURODA and MANUEL [15] recently pointed out that the enrichment of heavy xenon isotopes in carbonaceous and gas-rich meteorites is paralleled by an enrichment of heavy neon isotopes. These authors concluded that the enrichment of the heavier isotopes of both gases result from isotopic mass fractionation. Additional evidence for the production of isotopic anomalies of meteoritic xenon and krypton by fractionation have been reported from noble gas analyses of the LEOVILLE [21] and POTTER [22] chondrites.

This study of the isotopic composition of noble gases in ordinary chondrites was undertaken in order to obtain additional data with which to examine the current framework of ideas on the origin of noble gas anomalies in meteorites. Samples of three ordinary chondrites were used for this investigation.

Measurements

Relatively large meteorite samples, weighing about 10 grams each, were used in this study. These were purchased from the American Meteorite Laboratory (AML). The name and type of each meteorite, the sample weight and the AML catalogue number of the specimen from which our samples were taken are as follows: The Wellman, Texas olivine bronzite chondrite, 10.888 grams from specimen # H 12.56; the Tell, Texas olivinie hypersthene chondrite, 8.474 grams, from specimen # H 24.67; and the Scurry, Texas olivine bronzite chondrite, 7.909 grams, from specimen # H 65.25.

The samples were mounted in side-arm chambers of a quartz extraction bottle. The samples were heated to ≈ 60 °C overnight to remove surface contamination, the molybdenum crucible was degassed at 1800 °C and the pressure of the system reduced to 5×10^{-8} Torr. The extraction bottle and preliminary "getters" were then isolated from the rest of the system by metal bellows valves, the sample to be analyzed was then dropped magnetically into the molybdenum crucible which was heated by a radiofrequency induction heater. An optical pyrometer was used to estimate the extraction temperatures. Each extraction temperature was maintained for 30 minutes, and the evolved gases were cleaned in the extraction system by titanium at 850 °C and by copper-oxide at 550 °C. After these preliminary "getters" were cooled to room temperature, the gases were then pumped through the metal valve into a second clean-up system by adsorption on charcoal cooled witl liquid N_2 or solid CO_2. After isolating the gases in the second clean-up system, they were driven from the charcoal at 200 °C and exposed to a second surface of titanium sponge at 850 °C.

In the case of Wellman, the noble gases were separated into four fractions for analysis by adsorption on charcoal; the boiling temperature of N_2 released He and Ne, the sublimation temperature of CO_2 re-

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leased Ar, the freezing point of Hg released Kr, and finally the Xe was released from the charcoal at 150 °C. For Tell and Scurry the noble gases were pumped into the second clean-up system by adsorption on charcoal cooled with solid CO_2, and those gases not retained on charcoal at this temperature (He, Ne and Ar) were pumped from the second clean-up system prior to the final scrubbing on hot titanium (850 °C). The krypton and xenon were let into the mass spectrometer as separate fractions by selective adsorption of the xenon on charcoal at the freezing point of mercury.

The gases were analyzed statically in a Reynolds' type 4.5 inch 60° sector mass spectrometer [23]. The mass spectrometer was calibrated before and after each sample by analyzing small volumes of air ($\approx$0.01 cc STP) by the same procedure of analysis as used for the sample. The sensitivity of the spectrometer for each noble gas and the mass discrimination across the isotopes of each gas were calculated by comparing the peak heights with the atmospheric abundances of noble gases [24] and the isotopic composition of atmospheric neon [25], argon [26], krypton [27] and xenon [28]. No correction for mass discrimination was applied to helium. The errors reported in the isotope ratios are one standard deviation (σ) from the least squares line thorugh the observed ratios as a function of residence time in the mass spectrometer. Results from blank analyses, where the identical procedures were followed for gases evolved from a hot molybdenum crucible, showed no significant contamination except at mass 78. Due to variations in the sensitivity of the mass spectrometer the gas content has much larger errors than the isotope ratios, such that the content of each noble gas has an estimated error of $\pm 20\%$.

Results and Discussion

1. Noble Gases in Wellman

The abundances and isotopic composition of the noble gases in Wellman are shown in Table 1 together with the isotopic composition of trapped noble gases in carbonaceous chondrites and the solar-type noble gases observed in gas-rich meteorites and in the moon. The isotopic composition of trapped He, Ne and Ar in carbonaceous chondrites are the range of values reported in a comprehensive study [13, 14] and the trapped Kr and Xe are the average values [29, 30] observed in carbonaceous chondrites. The isotopic composition of solar-type He, Ne and Ar are from the Fayetteville gas-rich meteorite [7],

Table 1. Concentration and isotope composition of noble gases in the Wellman chondrite.

Noble gases	Wellman observed	Wellman trapped	Trapped gas in carbonaceous chondrites	Solar gas
He^3/He^4	0.0253 ± 0.0004	0.88×10^{-4}	$(1.25-4.20) \times 10^{-4}$	$\leqq 3.1 \times 10^{-4}$
He^4	6.04×10^{-7} cc STP/gm	5.25×10^{-7} cc STP/gm		3.08×10^6 per 10^6 atoms Si
Ne^{20}/Ne^{22}	1.415 ± 0.003	7.3	$7.77-13.10$	$\geqq 12.6$
Ne^{21}/Ne^{22}	0.876 ± 0.003	0.025	$0.025-0.036$	$\geqq 0.033$
Ne^{22}	5.39×10^{-8} cc STP/gm	4.35×10^{-9} cc STP/gm		8.36×10^5 per 10^6 atoms Si
Ar^{38}/Ar^{36}	0.450 ± 0.001	0.23	$0.172-0.200$	$\leqq 0.177$
Ar^{40}/Ar^{36}	3143 ± 26			
Ar^{36}	3.96×10^{-9} cc STP/gm	3.27×10^{-9} cc STP/gm		1.26×10^5 per 10^6 atoms Si
Kr^{80}/Kr^{84}	0.0423 ± 0.002	$\leqq 0.0423$	0.0392	0.0427
Kr^{82}/Kr^{84}	0.2048 ± 0.0007	0.2048	0.2012	0.2065
Kr^{83}/Kr^{84}	0.2045 ± 0.0006	0.2045	0.2016	0.2055
Kr^{86}/Kr^{84}	0.3058 ± 0.0007	0.3058	0.3097	0.297
Kr^{84}	1.41×10^{-10} cc STP/gm	1.41×10^{-10} cc STP/gm		2.93×10^1 per 10^6 atoms Si
Xe^{124}/Xe^{130}	0.0296 ± 0.0002	0.0296	0.0283	0.0299
Xe^{126}/Xe^{130}	0.0268 ± 0.0002	0.0268	0.0253	0.0288
Xe^{128}/Xe^{130}	0.508 ± 0.003	0.508	0.506	0.512
Xe^{129}/Xe^{130}	8.18 ± 0.05	(6.37)	(6.35)	6.38
Xe^{131}/Xe^{130}	4.97 ± 0.03	4.97	5.07	5.00
Xe^{132}/Xe^{130}	6.09 ± 0.03	6.09	6.21	6.05
Xe^{134}/Xe^{130}	2.36 ± 0.01	2.36	2.37	2.23
Xe^{136}/Xe^{130}	1.95 ± 0.01	1.95	1.99	1.81
Xe^{130}	1.17×10^{-11} cc STP/gm	1.17×10^{-11} cc STP/gm		1.62×10^{-1} per 10^6 atoms Si

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the Kr is from a lunar[31] breccia # 10021 and the solar Xe is that released from lunar[32] fines at 800 °C. The solar abundance of each rare gas is from the abundance tables of SUESS and UREY[33].

The isotopic composition of xenon and krypton in Wellman is intermediate between the isotopic composition of these gases in carbonaceous chondrites and in the moon. Except for the obvious excess of Xe^{129}, we interpret these two gases to represent the isotopic composition trapped in Wellman. The isotopic composition of the trapped component of the three light weight noble gases in Wellman is masked by a cosmogenic component. For helium and argon it is not possible to unambiguously separate the trapped and the cosmogenic components. However, these two components of neon can be separated by the method of MANUEL[7]. The concentration of trapped neon and its isotopic composition as calculated by this method are shown in Table 1. Since MANUEL[34] and SRINIVASAN and MANUEL[17] have shown that the He^3/He^4, Ne^{20}/Ne^{22} and Ar^{36}/Ar^{38} ratios in carbonaceous chondrites covary in the manner expected from a common mass-dependent fractionation process, we have shown in Table 1 the isotopic composition of trapped He and Ar obtained by assuming that the trapped He, Ne and Ar in Wellman have undergone a common fractionation process.

To compare the abundance of trapped noble gases in Wellman with the cosmic abundance of noble gases, we employ the equation of CANALAS et al.[3]

$$F^m = (X^m/Xe^{130})_{\text{sample}}/(X^m/Xe^{130})_{\text{cosmic}}, \quad (1)$$

where X^m is any trapped noble gas isotope with mass number m. Using the abundances of trapped

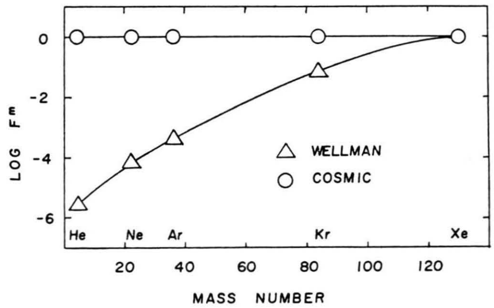

Fig. 1. Abundance pattern of noble gases in the Wellman chondrite relative to the cosmic abundances of these gases.

gases in Wellman from Table 1 and the cosmic abundances from SUESS and UREY[33], the fractionation pattern obtained for trapped noble gases in Wellman is shown in Fig. 1. Although the fractionation across the isotopes of a given noble gas cannot be quantitatively related to the separation of one gas from another since the latter may not depend on atomic mass alone[6], we note that the abundances of noble gases fit a smooth fractionation pattern and that this correlates with the difference between the isotopic composition of solar gases and the isotopic composition calculated for the trapped He, Ne and Ar in Wellman.

The fractionation pattern across the isotopes of trapped Kr and Xe in Wellman are shown in Fig. 2 and Fig. 3. The isotopic composition of Kr and Xe from average carbonaceous chondrites[29, 30] is shown for comparison. For these two plots Eq. (1) was modified to show the fractionation across trapped Kr and Xe by normalizing the mass spectrum to the heaviest isotope,

$$F^m_{Kr} = (Kr^m/Kr^{86})_{\text{sample}}/(Kr^m/Kr^{86})_{\text{solar}} \quad (2)$$

and

$$F^m_{Xe} = (Xe^m/Xe^{136})_{\text{sample}}/(Xe^m/Xe^{136})_{\text{solar}}. \quad (3)$$

In these equations the krypton from lunar breccia[31] # 10021 and the xenon released at 800 °C from lunar fines[32] # 10084 were used to represent solar gases. It has been noted earlier[22] that the possibility of gas loss and isotopic fractionation effects of the solar-type gases implanted on the lunar surface may correlate with long exposure ages and with a depletion of the light weight gases (He, Ne and Ar) relative to the heavier gases (Kr and Xe). These two analyses of lunar material were chosen to minimize these effects.

From Fig. 2 and Fig. 3 it can be seen that the preferential depletion of the lighter weight noble gases from Wellman, as shown in Fig. 1, has caused a selective depletion of the light isotopes of the heaviest noble gases, krypton and xenon. Furthermore, there is a correlation between the fractionation observed across the isotopes of these two noble gases: For both Kr and Xe the selective loss of the light isotopes is greater in average carbonaceous chondrites (AVCC)[29, 30] than in Wellman. The re-

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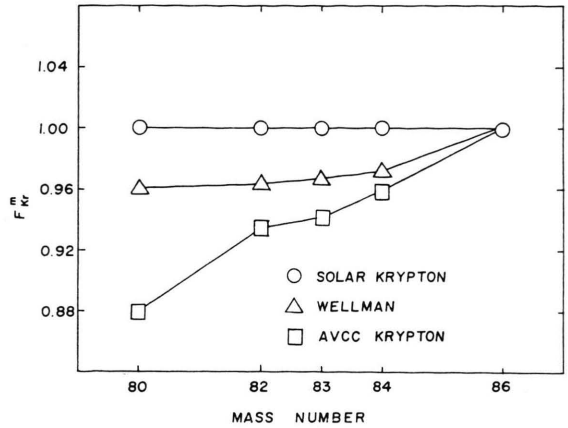

Fig. 2. Abundance pattern of krypton isotopes in the Wellman chondrite and in average carbonaceous chondrites relative to solar krypton in lunar breccia # 10021.

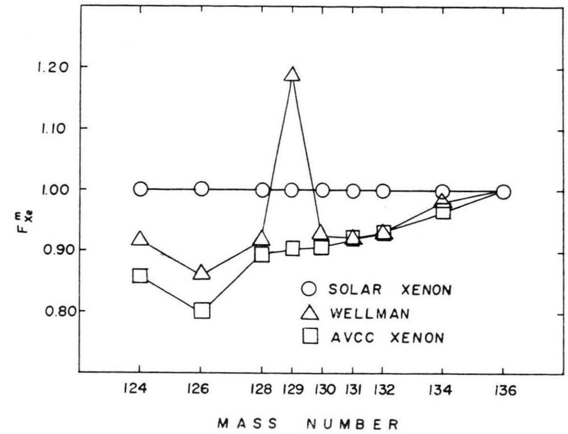

Fig. 3. Abundance pattern of xenon isotopes in the Wellman chondrite and in average carbonaceous chondrites relative to solar xenon released at 800 °C from lunar fines # 10084.

sults of this study on the isotopic composition of trapped xenon and krypton in Wellman therefore show evidence of a common fractionation process, as has been reported earlier in the Leoville carbonaceous chondrite [21].

The fine structure features of the fractionation curve shown in Fig. 3 is intriguing. KURODA [35] has considered this problem in detail and concluded that nuclear reactions which have occurred in the sun over geologic time may be responsible for some of these effects. Relative to solar xenon the minimum at Xe^{126} and the excess at Xe^{124} over that expected from fractionation could arise from a deficiency of Xe^{124} and an excess of Xe^{126} in the xenon currently implanted on the lunar surface from the solar wind. It is not unlikely that the abundance of these two xenon isotopes have been altered by nuclear reactions in the sun; Xe^{124} has the largest thermal neutron-capture cross section of any stable xenon isotope and the production of Xe^{126} from I^{127} via the (γ,n) reaction is ≈ 0.5 barns for high energy photons [36].

2. Krypton and Xenon in Tell and Scurry

The abundance and isotopic composition of xenon and krypton from the stepwise heating of Tell and Scurry are shown in Table 2 and Table 3, respectively.

In Tell the atomic weight of both xenon and krypton is lightest in the gases released at the highest extraction temperature. Although there is evidence

Table 2. Concentration and isotopic composition of krypton and xenon released by stepwise heating of the Tell chondrite.

Noble gas	Extraction Temperature 600°	900°	Melt	Solar gas
Kr^{80}/Kr^{84}	0.0384 ± 0.0004	0.0400 ± 0.0009	0.0452 ± 0.0006	0.0427
Kr^{82}/Kr^{84}	0.199 ± 0.001	0.202 ± 0.002	0.204 ± 0.001	0.2065
Kr^{83}/Kr^{84}	0.199 ± 0.001	0.200 ± 0.002	0.205 ± 0.001	0.2055
Kr^{86}/Kr^{84}	0.306 ± 0.001	0.306 ± 0.002	0.307 ± 0.001	0.297
$Kr^{84} \times 10^{-10}$ cc STP/gm	0.80	0.35	0.33	
Xe^{124}/Xe^{130}	0.0242 ± 0.0004	0.0229 ± 0.0006	0.0292 ± 0.0003	0.0299
Xe^{126}/Xe^{130}	0.0227 ± 0.0004	0.0236 ± 0.0005	0.0275 ± 0.0004	0.0288
Xe^{128}/Xe^{130}	0.477 ± 0.004	0.473 ± 0.006	0.509 ± 0.005	0.512
Xe^{129}/Xe^{130}	6.50 ± 0.06	6.62 ± 0.04	7.68 ± 0.07	6.38
Xe^{131}/Xe^{130}	5.19 ± 0.04	5.21 ± 0.04	5.03 ± 0.03	5.00
Xe^{132}/Xe^{130}	6.57 ± 0.05	6.62 ± 0.03	6.16 ± 0.03	6.05
Xe^{134}/Xe^{130}	2.53 ± 0.01	2.57 ± 0.01	2.38 ± 0.02	2.23
Xe^{136}/Xe^{130}	2.16 ± 0.01	2.19 ± 0.02	2.00 ± 0.02	1.81
$Xe^{130} \times 10^{-12}$ cc STP/gm	5.3	1.2	7.8	

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Table 3. Concentration and isotopic composition of krypton and xenon released by stepwise heating of the Scurry chondrite.

Noble gas	Extraction Temperature 600°	900°	Melt	Solar gas
Kr^{80}/Kr^{84}	0.0396 ± 0.0005	0.0397 ± 0.0003	0.0420 ± 0.0003	0.0427
Kr^{82}/Kr^{84}	0.201 ± 0.001	0.198 ± 0.001	0.204 ± 0.001	0.2065
Kr^{83}/Kr^{84}	0.200 ± 0.002	0.198 ± 0.001	0.204 ± 0.001	0.2055
Kr^{86}/Kr^{84}	0.306 ± 0.001	0.306 ± 0.001	0.306 ± 0.001	0.297
$Kr^{84} \times 10^{-10}$ cc STP/gm	0.48	0.79	2.2	
Xe^{124}/Xe^{130}	0.0256 ± 0.0005	0.0244 ± 0.0002	0.0251 ± 0.0002	0.0299
Xe^{126}/Xe^{130}	0.0226 ± 0.0005	0.0234 ± 0.0004	0.0242 ± 0.0003	0.0288
Xe^{128}/Xe^{130}	0.479 ± 0.003	0.488 ± 0.003	0.493 ± 0.004	0.512
Xe^{129}/Xe^{130}	6.58 ± 0.04	6.69 ± 0.03	6.69 ± 0.03	6.38
Xe^{131}/Xe^{130}	5.17 ± 0.04	5.19 ± 0.03	5.11 ± 0.02	5.00
Xe^{132}/Xe^{130}	6.53 ± 0.05	6.59 ± 0.05	6.40 ± 0.02	6.05
Xe^{134}/Xe^{130}	2.51 ± 0.01	2.55 ± 0.02	2.46 ± 0.01	2.23
Xe^{136}/Xe^{130}	2.14 ± 0.01	2.14 ± 0.01	2.07 ± 0.01	1.81
$Xe^{130} \times 10^{-12}$ cc STP/gm	2.4	3.6	7.6	

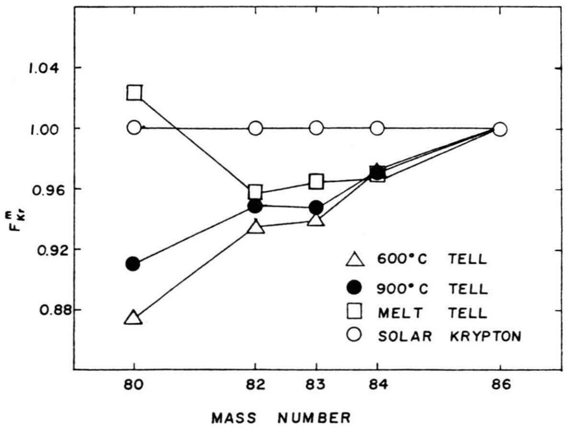

Fig. 4. Abundance pattern of krypton isotopes in the different temperature fractions of the Tell chondrite relative to solar krypton.

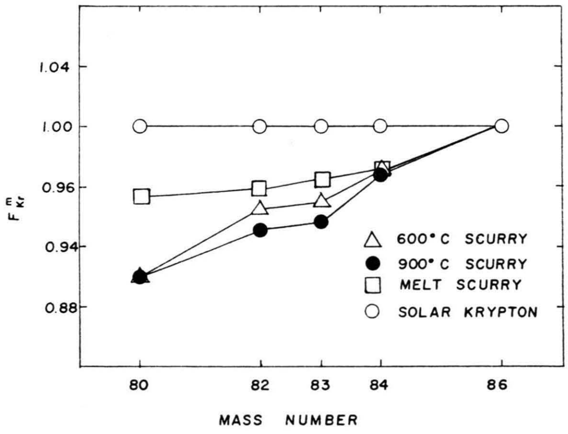

Fig. 6. Abundance pattern of krypton isotopes observed by stepwise heating of the Scurry chondrite normalized to solar krypton.

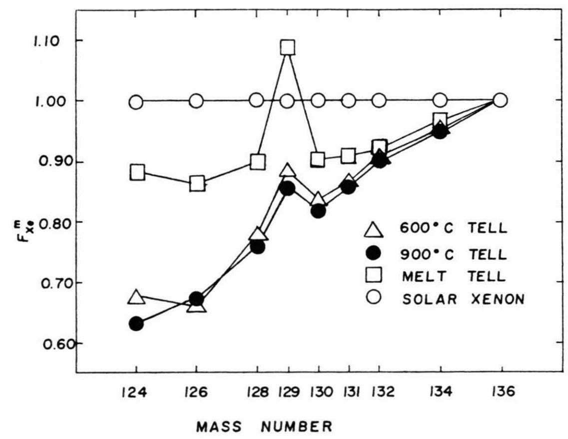

Fig. 5. Abundance pattern of xenon isotopes in the different temperature fractions of the Tell chondrite relative to solar xenon.

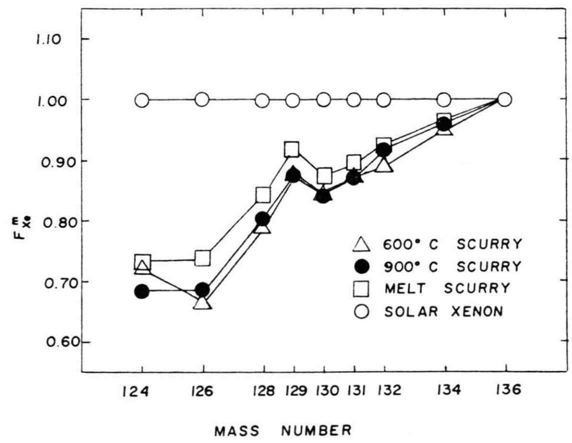

Fig. 7. Abundance pattern of xenon isotopes observed by stepwise heating of the Scurry chondrite normalized to solar xenon.

for small excesses of Kr^{80}, Xe^{124} and Xe^{126} from spallation in the melt fraction of Tell, isotopic fractionation plus the *in situ* decay of I^{129} seem to be the two dominant effects in generating variations in the isotopic composition of Kr and Xe. This is shown in Fig. 4 and Fig. 5, where the Kr and Xe isotope ratios from the stepwise heating of Tell are shown in the manner employed for Wellman.

The isotopic composition of Kr and Xe released by stepwise heating of Scurry shows the same general trend as the gases in Tell. This is shown in Fig. 6 and Fig. 7, where again we observe a covariance in the fractionation across the isotopes of Kr and Xe. In both Tell and Scurry there is a small excess of Kr^{80}, which we attribute to spallation reactions [37] induced by cosmic rays. The less pronounced minimum at Xe^{126} in Tell and Scurry (Fig. 5 and Fig. 7) than in Wellman and AVCC (Fig. 3) is also suggestive of a small component of spallation-produced [37] Xe^{126} in the xenon of Tell and Scurry.

Since the xenon and krypton released in different temperature fractions of the Scurry and Tell chondrites show variations due to mass fractionation, it seems likely that mass fractionation has also played a role in generating some of the isotopic variations of noble gases reported in the different temperature fractions of carbonaceous and gas-rich chondrites [7, 10, 11, 21, 38]. Evidence for this process is compelling in view of (a) the covariance of the isotopic composition of xenon with krypton in the ordinary chondrites studied here and in carbonaceous chondrites [21], (b) the covariance of trapped xenon with trapped neon in the different temperature fractions of carbonaceous and gas-rich meteorites [15], (c) the covariance of He^3/He^4, Ne^{20}/Ne^{22} and Ar^{36}/Ar^{38} for the total trapped gas in carbonaceous chondrites [17], and (d) the general agreement of the isotopic fractionation pattern observed across each noble gas with the fractionation pattern observed across the abundances of all the trapped noble gases.

Thus, there is cogent evidence of a mass dependent fractionating process which altered both the abundance pattern of noble gases and their isotopic composition in planetary material. It should be emphasized that the isotopic fractionation of noble gases has been known for years. In 1913 ASTON [39] obtained evidence for isotopes by altering the density of neon by diffusion through clay pipe stems. It appears that solid planetary material has behaved as the clay stems in selectively retaining the heavier isotopes of the noble gases.

Acknowledgements

We are grateful to Mr. J. V. ALBRECHT for assistance with data reduction and to Mr. B. SRINIVASAN and Mr. D. E. SINCLAIR for many stimulating discussions. This work was supported by the U.S. National Science Foundation, Grant NSF-GA-16618.

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