

Thermal metamorphism of primitive meteorites—VIII. Noble gases, carbon and sulfur in Allende (C3) meteorite heated at 400–1000°C

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Abstract—Noble gases, C and S are lost from Allende samples heated for 1 week at temperatures of 400–1000°C in a low pressure environment. In the extreme, losses of ^3He and ^4He are $\sim 100\times$ while for C, S and Ne, Ar and Kr isotopes and ^{132}Xe , these are $\leq 10\times$. Except for He, these losses are less severe than those of Bi or Tl from samples heated in the same runs. Significant He, Ne and Ar isotopic fractionation during heating indicates preferential outgassing of specific reservoirs. Apparent activation energies for all species generally indicate loss controlled by a diffusive process. Next to He, ^{40}Ar is the most labile of those species considered here but still less so than Bi or Tl. L-group (but not H- or LL-group) chondrites may have lost mobile elements like Tl while being outgassed after late impact-associated heating. A less likely alternative possibility involving a collateral relation between condensation conditions and depth in a parent object may also explain the L-group trend.

INTRODUCTION

IN PAPER I of this series, IKRAMUDDIN and LIPSCHUTZ (1975) demonstrated mobilization, i.e. volatilization and loss, of certain trace elements from chondritic material by week-long heating under ambient conditions reasonable for primitive solar system objects during thermal metamorphism. The conditions used in Paper I—i.e. the heating of aliquots at 100°C increments at 400–1000°C at low pressures (initially $\sim 10^{-5}$ atm H_2)—remained standard for subsequent studies. Paper I involved six trace elements—Bi, Co, Ga, In, Se and Tl—in the Allende (C3) chondrite, all but Co being mobile to some extent at least. These and other mobile elements (Ag, Cd, Cs, Te and Zn) were studied in samples as compositionally and genetically diverse as primitive ordinary chondrites, a terrestrial basalt, C2 and E4 chondrites (IKRAMUDDIN *et al.*, 1976a,b, 1977a,b; MATZA and LIPSCHUTZ, 1977a—hereafter referred to as Papers II–IV, respectively). The trace element retention patterns are surprisingly similar although differences in detail are apparent. Similarities between trace element trends in heated Abee (E4) with those in E4-6 chondrites are consistent with the suggestion that E-group chondrites experienced substantial open-systems, thermal metamorphism (Papers II, VI; cf. BINZ *et al.*, 1974).

Differences between trace element trends in heated Tieschitz (H3) and Krymka (L3) and in H3-6 and L3-6 chondrites, respectively, suggest that ordinary chondrites escaped such metamorphism (Papers III, V; cf. BINZ *et al.*, 1976) with their compositions apparently reflecting condensation [cf. ANDERS *et al.* (1976), BINZ *et al.* (1976) and earlier references cited in these papers]. C3-4 chondrites present a less-definitive picture (MATZA and LIPSCHUTZ, 1977b, 1978, work in progress).

In papers I–VI, we chose to study a suite of elements expected to be relatively mobile during thermal metamorphism—and to compare the response in artificially heated materials with trends observed in more evolved meteorites. In all experiments we study elemental mobility rather than volatility; volatility controls an element's distribution between gas and solid at local thermodynamic equilibrium. Experimental constraints prevent attainment of equilibrium so we refer to the observed tendency of an element to vaporize and be lost, as its mobility. We assume that mobility trends in our experiments parallel volatility trends in actual metamorphism in parent bodies. This approach may be superior to a direct calculation of volatility based on thermodynamic data or estimated in other ways. Direct calculations are difficult because trace elements do not generally form discrete chemical compounds, i.e. minerals, with known thermodynamic properties. Indeed, they may not even be

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sited in a single mineral but rather may be located in or on numerous such sites.

In this study we enlarge the coverage of volatile elements to include the noble gases, carbon and sulfur. By any criterion, noble gases should be among the most volatile elements. Carbon was included because of its large chondritic depletion factor (VDOVYKIN and MOORE, 1971)—i.e. the ratio of its abundance in ordinary chondrites, especially in petrologic types 5 and 6, relative to its cosmic or CI abundance (cf. BINZ *et al.*, 1976). The depletion factor provides a qualitative measure of elemental volatility during nebular condensation (ANDERS, 1964) but not necessarily under other conditions. Carbon should be especially volatile in carbonaceous chondrites in which it exists principally in organic compounds (cf. VDOVYKIN, 1967); in Allende it probably exists as a thin-film coating on mineral grains (CLARKE *et al.*, 1970), possibly as amorphous carbon (LEWIS, personal communication). Stepwise heating has long been used (e.g. ZÄHRINGER, 1962) to characterize meteoritic noble gas components with differing isotopic compositions, hence differing nucleosynthetic, fissionogenic, radiogenic or cosmogenic origins [cf. MANUEL *et al.* (1972), SMITH *et al.* (1977) and SRINIVASAN *et al.* (1978) for examples of such studies of Allende]. However such experiments differ from the integral release measurements of Papers I–VI. Step-wise heating involves differential gas release in relatively short heating times of ~1 hr and extraction into very high vacuum, $\leq 10^{-6}$ torr, in the presence of various metal getters. These conditions are so different from those of Papers I–VI that direct comparison of noble gas data from differential experiments with other mobile elements in integral experiments is of dubious value.

For comparative purposes we felt it worthwhile analyzing noble gas retention in a chondrite heated under our standardized conditions and, for several reasons, chose Allende. First, when current experiments are complete, many more elements will have been determined in Allende than in any other heated chondrite; a complete mineralogic–petrologic description of heated samples will also be reported. Second, we anticipated that noble gases would be extremely mobile and that their low concentrations in high-temperature samples might make measurement difficult. Allende seems the most retentive chondrite studied to date (Papers I–VI) thus minimizing this problem. Finally, as noted there is a considerable body of high-precision noble gas data for Allende obtained from single- and multi-step heating experiments with which to compare our results. Since C and, to a lesser extent, S were also expected to be rather mobile we decided to measure these on the same aliquots. We report the data for these two elements and the noble gases here. In discussing these results we compare general trends for these elements and those determined previously (Paper I; IKRAMUDDIN *et al.*, 1975). We reserve detailed comparison pending completion of the chemical, mineralogic and petrologic studies.

EXPERIMENTAL

All measurements were carried out on aliquots of homogenized Allende powder from USNM-3643 heated in the same runs that provided samples for trace element analysis (cf. Paper I; IKRAMUDDIN *et al.*, 1975). The aliquots still sealed in thin quartz vials, were sent to E.K.G. who removed samples for C and S determination by the techniques of MOORE *et al.* (1970) and GIBSON and MOORE (1974). The material remaining was then sent to G.F.H. for mass spectrometry. Unfortunately, the post-heating weight of the entire aliquot was not recorded so that C, S and noble gas concentrations in this paper are on a volatile-free basis. This additional uncertainty should have little or no effect upon our conclusions since the weight loss on heating C3 chondrites to temperatures as high as 1300°C is but 4–8% (GIBSON *et al.*, 1974).

All mass spectrometric measurements were carried out at the Max-Planck-Institut für Kernphysik in Heidelberg using conventional techniques. Samples were pre-heated for 24 hr at 120°C prior to vaporization at 1650°C. The gases were cleaned by successive 15-min exposures to hot Ti getters (850°C), after which Ar, Kr and Xe were adsorbed on a charcoal cold-finger and He and Ne were admitted to the spectrometer for measurement. After isolating the sample system, the heavy gases were released at 20°C and Xe and nearly all of the Kr were reabsorbed on a glass U-tube (–196°C). Ar was admitted to the spectrometer and the amount of ^{84}Kr monitored; this amount was always small, i.e. <4% of the total Kr admitted to the spectrometer together with Xe after warming the U-tube to 20°C. The Xe spectrum was scanned at least ten times by a computer-controlled peak jumping program; no systematic time dependence of the Xe isotopic ratios was observed. Other mass ranges were scanned manually and when necessary the results were extrapolated linearly to the time of admission to the spectrometer.

The clean-up procedures for the daily blanks and periodic standards were less stringent than those for samples but a careful comparison of the full and abbreviated procedures for one standard and one blank revealed no significant difference. Over the 4-week course of the measurements the machine sensitivity was stable to within 5% in all cases.

It became apparent after the first bulk sample measurement that the He–Ne–Ar standard had been pumped inadvertently at some earlier time. This standard was therefore re-calibrated against a second one attached to the same machine (T. KIRSTEN, personal communication), with a stated accuracy of $\pm 5\%$ (HAAG, 1975). For our purposes the ratios of gas contents before and after heating are of primary importance and the uncertainty introduced here is of minor importance.

The operating conditions for the spectrometer were: ion acceleration, 2600 V; electron acceleration, 40 V; electron multiplier, 3000 V; filament current, 30 μA .

RESULTS AND DISCUSSION

Unheated samples: accuracy and precision

The C and S results for heated samples are listed in Table 1 together with those for an unheated aliquot of the same batch and another batch of Allende reference standard. The C content of the first unheated sample is similar to values reported previously—2700 ppm (KING *et al.*, 1969) and duplicates of 2900 ppm (CLARKE *et al.*, 1971). These are substantially lower than the value for the second unheated sample suggesting some problem with this sample; this conclusion is reinforced by data for heated samples (Table 1). Only those samples heated at $\geq 700^\circ\text{C}$ show unambiguous C loss; the 4 remaining heated samples

Table 1. Carbon and sulfur contents of unheated and heated Allende

Sample (°C)	Carbon [¶] (ppm)	Sulfur [§] (%)
unheated*	2820±140	2.51±0.12
unheated	3640±200	2.44±0.12
400	2750±140	2.44±0.12
500 [§]	<i>3010±150</i>	<i>2.49±0.12</i>
	3110±150	2.40±0.12
600	2780±140	2.33±0.11
700	1950±100	2.43±0.12
800	1430±80	2.28±0.11
900	990±50	2.27±0.11
1000	1050±50	1.73±0.08
(He) [†]	<i>1180±80</i>	<i>1.98±0.10</i>
(O ₂) [†]	<i>1040±50</i>	<i>2.07±0.10</i>

* Data are for another homogenized Allende reference sample (sample 10, split 13) obtained from the U.S. National Museum and are listed for comparative purposes only.

§ Data in italics are for a powder sample heated for 29 days (cf. IKRAMUDDIN *et al.*, 1975).

† Data in italics are for powder samples in which the initial ambient atmospheres were $\sim 10^{-3}$ atm of He or O₂ (cf. IKRAMUDDIN *et al.*, 1975).

¶ Each datum represents a single analysis. The uncertainties quoted are $\pm 5\%$ for C and S; these are reasonable, even conservative, limits in the light of previous studies (see text) which generally involve C and S contents lower than those reported here.

have a mean content of 2910 ± 180 ppm. To be conservative, we take this as the value for unheated Allende from this batch of reference standard. However, our conclusions are generally unaffected by this choice.

The S data (Table 1) indicate systematic bias. Values for both unheated samples and the three heated at $\leq 500^\circ\text{C}$ are very similar (to within 2%); these results and, indeed, nearly every S datum in Table 1 exceed the values of 2.06 and 2.07% determined by wet chemistry (CLARKE *et al.*, 1971). We have no particular reason to doubt the accuracy of our measurements. In lunar samples, which are an order of magnitude or more lower in S than Allende, results obtained by acid hydrolysis (e.g. KAPLAN *et al.*, 1976) or X-ray fluorescence (RHODES *et al.*, 1976) are 10–30% lower than those determined by combustion in O₂ (e.g. GIBSON and MOORE, 1974; GIBSON *et al.*, 1976). These lower results apparently result from incomplete dissolution of S-bearing phases during acid hydrolysis (cf. KERRIDGE *et al.*, 1975; DES MARAIS, personal communication) and partial vaporization during preparation of fused beads for X-ray fluorescence analysis. In any event, for our purposes accuracy does not play as great a role as precision and we are satisfied that the precision of our measurements is entirely satisfactory.

In Table 2 we list noble gas results for unheated and heated aliquots of the Allende reference standard and for an unheated chip from Me-2639 obtained from the Field Museum. All data are corrected for blanks, mass discrimination and time-dependent variations; we also list typical blank values. For brevity we list only ⁸⁴Kr and ¹³²Xe in Table 2 although we determined all other Kr and Xe isotopes as well. Overall trends for ⁸⁴Kr and ¹³²Xe were similar to those for other Kr and Xe isotopes, respectively and the isotopic ratios neither varied systematically with temperature nor permitted conclusions concerning detailed isotopic trends. Depending upon the specific ratio con-

Table 2. Noble gas retentivities in Allende samples

Sample	Wt(mg)	Noble Gas Concentrations*									
		³ He	⁴ He	²⁰ Ne	²¹ Ne	²² Ne	³⁶ Ar	³⁸ Ar	⁴⁰ Ar	⁸⁴ Kr	¹³² Xe
unheated [§]	16.4	10.1± 0.3	3720± 150	7.71± 0.67	2.32± 0.07	3.29± 0.14	17.4	3.23	2070	1490	1200
unheated	9.1	8.0± 0.4	3790± 170	5.2± 1.0	2.43± 0.09	3.02± 0.16	16.1	2.96	2640	----	----
400°C	27.5	5.0± 0.2	2620± 100	7.89± 0.39	2.44± 0.07	3.39± 0.13	18.7	3.58	2000	1790	1460
500°C [†]	17.1	2.3± 0.1	<i>3640± 120</i>	<i>6.22± 0.58</i>	<i>2.15± 0.07</i>	<i>3.93± 0.13</i>	<i>18.1</i>	<i>3.46</i>	<i>1355</i>	<i>1880</i>	<i>1340</i>
	41.1	2.6± 0.1	1610± 120	6.16± 0.31	1.93± 0.06	2.48± 0.09	15.5	3.13	1170	1640	1410
600°C	36.8	1.4± 0.1	1070± 40	4.83± 0.32	1.94± 0.06	2.52± 0.09	18.5	3.54	1180	1810	1520
700°C	59.9	0.43± 0.02	555± 20	4.39± 0.21	1.69± 0.06	2.24± 0.08	19.1	3.63	925	1610	1450
800°C	46.3	0.10± 0.01	170± 7	2.56± 0.24	1.31± 0.05	1.61± 0.07	17.8	3.41	665	----	----
900°C	48.2	0.03± 0.01	95± 5	1.74± 0.18	0.91± 0.03	1.12± 0.05	13.2	2.55	415	1155	950
1000°C [†]	31.3	----	26± 5	0.84± 0.24	0.50± 0.02	0.63± 0.05	8.2	1.62	485	605	525
(O ₂)	48.1	----	28± 5	1.05± 0.17	0.61± 0.02	0.72± 0.04	7.8	1.52	400	655	525
blank [¶]	----	0.2	120	9	0.05	1.0	0.6	0.2	34	3	0.6

* Concentration units are 10^{-8} cm³ (STP)/g for all gases but ⁸⁴Kr and ¹³²Xe which are 10^{-12} cm³ (STP)/g. We estimate 1 σ uncertainties of 7–8% for Ar isotopes, ⁸⁴Kr and ¹³²Xe from the dispersion of the individual measurements for our unheated sample and those heated at $\leq 700^\circ\text{C}$. Slightly lower limits ($\pm 5\%$) would have been estimated from consideration of all sources of error except the absolute uncertainty of the standard; we believe the uncertainties quoted should be used in comparing the gas contents measured in different runs.

§ Data for a chip from Me-2639 for comparative purposes only.

† Data in italics indicate non-standardized heating conditions: 4 weeks' heating time at 500°C and an initial pressure of $\sim 10^{-3}$ atm O₂ at 1000°C.

¶ Units for blank are 10^{-10} cm³ (STP) for all gases but ⁸⁴Kr and ¹³²Xe which are 10^{-13} cm³ (STP).

sidered. Kr isotopic ratios (relative to ^{84}Kr) varied by 1–7% and Xe isotopic ratios (relative to ^{132}Xe) varied by 2–9% over the entire temperature range. Tables containing these data and prior whole-rock literature data for Allende are available from G.F.H. or M.E.L.

Our data for unheated samples or the sample heated at 400°C appear normal but a bit higher than literature values. The Ar data of MANUEL *et al.* (1972) are definitely discrepant but at this late date it appears impossible to establish the cause for this (MANUEL, personal communication). Unfortunately, these are the only Ar literature values obtained by step-wise heating of bulk Allende. Were they valid, they would be valuable for comparison with our own data. The $^{36}\text{Ar}/^{38}\text{Ar}$ and $^{40}\text{Ar}/^{38}\text{Ar}$ ratios of MANUEL *et al.* (1972) are bracketed by those of MAZOR *et al.* (1970) and LEWIS *et al.* (1977) and are similar to those of FIREMAN *et al.* (1970).

In most of the following discussion, absolute noble gas contents are unimportant; only post-heating concentrations in samples relative to initial values are of interest. In contrast to previous practice for other elements (Papers I–IV) we chose to normalize noble gas contents to those in the sample heated at 400°C rather than in unheated Allende. Unheated samples were somewhat smaller (Table 2) and were run under slightly different conditions from heated ones. Furthermore, the Kr fraction of the unheated sample was lost and, for consistency, it seemed best to treat all noble gases similarly. The use of the 400°C sample as a standard has additional advantages of minimizing the effect of reporting data on a volatile-free basis and permitting compensation for adsorbed atmospheric constituents that might not be desorbed at 120°C (cf. ZÄHRINGER, 1962). In fact, this choice does not modify our conclusions significantly.

Elemental isotopic retentivities

The C and S contents of heated samples indicate incipient loss at 700 and 800°C, respectively (Table 1). Carbon concentrations generally decrease progressively with temperature, an unexpectedly-high ~30% of the initial C being retained even at 1000°C. In contrast, S losses at 800 and 900°C are marginal and even at 1000°C, 70% of the initial S is retained. To some extent this resembles the trend of Se in Allende aliquots heated in the same runs (Paper I) suggesting that the mobile portions of each element are sited in the same location(s). This conclusion is supported by the near-constancy of S/Se weight ratios and the significant S–Se correlation in unheated and heated

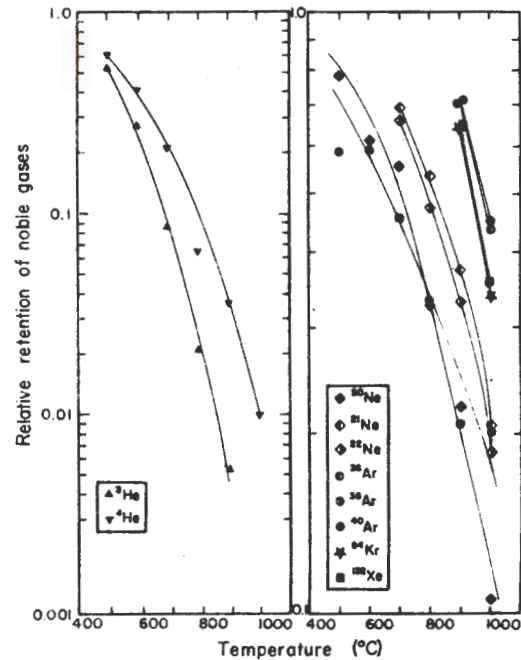


Fig. 1. Retention of noble gases in Allende as a function of heating temperature. Data are included only if unambiguous loss occurred at that temperature. The curves shown are visual fits to the data. The small overall variation of the Ne and Ar isotopes, ^{84}Kr and ^{132}Xe is noteworthy, these data generally varying by $<10\times$ overall. Over the same temperature range, elements which are not gases at room temperature—e.g. Bi, In or Tl—are more mobile in Allende (see text).

Allende: 2600 ± 200 for samples heated under standardized conditions and 2700 ± 400 for all samples ($>96\%$ and $>93\%$ confidence levels, respectively). Only the 1000°C samples show any evidence for fractionation (S/Se of 2200), but this is marginal at best. The constancy of chondritic S/Se ratios has been discussed previously (PELLY and LIPSCHUTZ, 1971) particularly with reference to S/Te and Se/Te fractionation in E3-6 and, perhaps, C3 chondrites (cf. BINZ *et al.*, 1974; Paper II). The relatively high S retention even at 1000°C seems especially surprising since IKRAMUDDIN *et al.* (1975) could discern no visible sulfides in Allende samples heated at this temperature. These results suggest a host-site change for most of the S during heating with the non-mobile 2/3 changing its chemical bonding to alternate sites, perhaps in the matrix as in similarly-heated Murchison (MATZA and LIPSCHUTZ, 1978). Mineralogic/petrologic study of heated Allende should shed further light on this question (BART and LIPSCHUTZ, in progress).

Noble gas retention patterns derived from data in Tables 2 and 3 are illustrated in Fig. 1. As for other mobile elements (Papers I–VI) losses of these gases progress with temperature and occur over a broad temperature span. As expected, He isotopes are the most mobile, being lost at temperatures of $\geq 500^\circ\text{C}$ and varying by factors ≥ 100 at temperatures up to 1000°C. Patterns for other noble gas nuclides are more interesting because of their high degree of reten-

Table 3. Apparent activation energies for loss of noble gas isotopes and carbon from Allende

Nuclide or Element	Temp. Range (°C)	Q (kcal/mol)	Temp. Range (°C)	Q (kcal/mol)
^3He			500–900	13
^4He			500–1000	13
C			700–1000	19
^{20}Ne	800–1400	19*	500–1000	13
	600–1400	25†		
^{21}Ne	800–1400	23*	700–1000	20
	600–1400	21†		
^{22}Ne	800–1400	21*	700–1000	18
	600–1400	21†		
^{36}Ar	1000–1400	55*	900–1000	45
^{38}Ar	1000–1400	55*	900–1000	45
^{40}Ar	1000–1400	19*	500–1000	8.1
^{84}Kr			900–1000	45
^{132}Xe			900–1000	44

* Based upon data of MANUEL *et al.* (1972).

† Based upon data of SRINIVASAN *et al.* (1978).

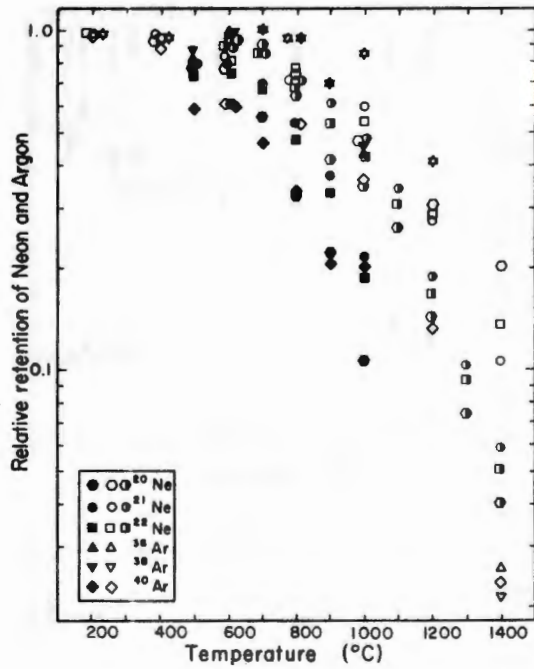


Fig. 2. Comparison of Ne and Ar isotopic retention in heated Allende samples. Results from step-wise heating experiments by MANUEL *et al.* (1972) and SRINIVASAN *et al.* (1978)—open and half-filled symbols, respectively—have been recalculated as integral release data for comparison with our experiments (filled symbols). The clots at 200 and 400 C include both sets of literature values. These results indicate that ^{20}Ne and ^{40}Ar are generally more mobile than $^{21,22}\text{Ne}$ and $^{36,38}\text{Ar}$, respectively (see text).

tion and substantial differences in the behavior of Ne and Ar isotopes. Except for He, losses at 1000°C are less than a factor of 10 whereas other mobile elements—e.g. Bi, In or Tl—in Allende powder aliquots heated in the same runs vary by at least this amount (Paper I). ^{20}Ne and ^{40}Ar are clearly more mobile than $^{21,22}\text{Ne}$ and $^{36,38}\text{Ar}$, both with respect to temperature

where losses exceed 10% and extent of loss over the entire temperature span. Both ^{20}Ne and ^{40}Ar are mobilized to a significant extent at $\geq 500^\circ\text{C}$; comparable losses of other Ne isotopes occur at $\geq 700^\circ\text{C}$ while $^{36,38}\text{Ar}$, like ^{84}Kr and ^{132}Xe , are appreciably mobilized only $\geq 900^\circ\text{C}$ (Fig. 1). By 1000°C, the contents of ^{20}Ne and ^{40}Ar are reduced to 10 and 20% of their initial values, respectively; 20% of the $^{21,22}\text{Ne}$ and 35–45% of the $^{36,38}\text{Ar}$, ^{84}Kr and ^{132}Xe each remain. [At the suggestion of a reviewer we emphasize that the concentration of a given noble gas isotope reflects contributions from radiogenic, cosmogenic and trapped components.]

Some of these trends appear in results from step-wise heating experiments on Allende. In Fig. 2 we depict our Ne and Ar results (Table 2) together with those calculated from data of MANUEL *et al.* (1972) and SRINIVASAN *et al.* (1978) by treating published results as integral release experiments. At temperatures $\geq 900^\circ\text{C}$, data of SRINIVASAN *et al.* (1978) indicate ^{20}Ne to be consistently more mobile than $^{21,22}\text{Ne}$. Results of MANUEL *et al.* (1972) yield no coherent trend for Ne but show ^{40}Ar to be lost preferentially at temperatures as low as 600°C. Because we heated our samples 100 times longer than the typical step of a stepwise heating experiment, it is only reasonable that retentions derived from literature data at a given temperature are considerably higher than those we determined; our results at 1000°C correspond roughly to samples run in step-wise fashion at 1200–1300°C (Fig. 2).

Isotopic ratios

It is well known that step-wise heating of meteorites leads to selective outgassing of distinct gas reservoirs with characteristic isotopic ratios. We depict the $^3\text{He}/^4\text{He}$ variation in two ways: as a function of the depletion of ^3He and of elemental He which for all practical purposes is ^4He (Fig. 3). In both cases $^3\text{He}/^4\text{He}$ ratios correlate with retention on linear

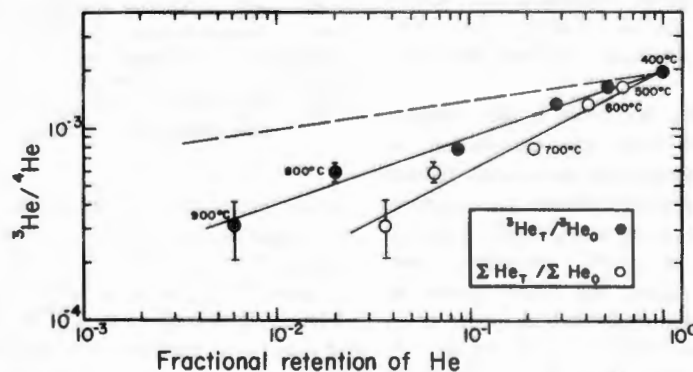


Fig. 3. He isotopic composition as a function of heating temperature, hence He retention. Closed symbols designate fractional retention of ^3He after heating at temperature T relative to that in Allende heated at 400°C; open symbols designate fractional retention of total He (which is, effectively, ^4He) in the same manner. The dashed line designates the He isotopic ratio generated by mass-fractionation during loss of He. Apparently, reservoirs of different isotopic compositions are being emptied during He loss.

regression, the correlation coefficients of 0.99 and 0.98, respectively, indicating significance at >99.99% confidence level for six samples. This would not be expected if rapid melting or decomposition of one or two well-defined minerals dominated the release of helium. Mass-fractionation can be ruled out based on a comparison with the line calculated from the equation of ASTON (1933),

$$\log R = \frac{(m_2 - m_1)}{(m_2 + m_1)} \cdot \log(C_0/C) \quad (1)$$

where R is the relative enrichment of the heavier isotope, m_2 , resulting from reduction of the initial gas concentration, C_0 , to a new value, C , after heating at a given temperature (cf. MANUEL *et al.*, 1972). [This equation yields the same relationship between $^3\text{He}/^4\text{He}$ and C/C_0 for He as that given by ZÄHRINGER (1962).] The data suggest preferential outgassing of two reservoirs with different origins, hence different proportions of ^3He and ^4He ; the lability of cosmogenic He apparently exceeds that of radiogenic ^4He .

Neon isotopic ratios are phenomenologically similar (Fig. 4a). Correlation coefficients for $^{20}\text{Ne}/^{22}\text{Ne}$ and $^{21}\text{Ne}/^{22}\text{Ne}$ vs Ne retention are 0.94 and 0.84, respectively, for 9 samples leading to significance levels of >99.99 and >99.7%, respectively. In the normal 3-isotope plot of $^{20}\text{Ne}/^{22}\text{Ne}$ vs $^{21}\text{Ne}/^{22}\text{Ne}$ the data array progresses almost monotonically with temperature toward the cosmogenic region— $^{20}\text{Ne}/^{22}\text{Ne} \sim 0.9$, $0.8 < ^{21}\text{Ne}/^{22}\text{Ne} < 1.0$. These data are consistent with trends in Fig. 5 of SMITH *et al.* (1977) showing that cosmogenic Ne is released preferentially from Allende matrix and aggregate samples at higher temperatures than trapped Ne. In C1 and C2 chondrites this trend is reversed: cosmogenic Ne tends to outgas at lower temperatures (BLACK, 1972; JEFFERY and ANDERS, 1970; HERZOG and ANDERS, 1974). This effect may be understood in terms of grain size variations (SMITH *et al.*, 1977). The accurate inference of trapped Ne ratios from our data is impeded by the dominance of cosmogenic Ne in the samples and the (unknown) variability of the cosmogenic Ne ratios. The Ne isotopic ratios obtained by step-wise heating by MANUEL *et al.* (1972) show no systematic variation with temperature.

^{38}Ar remains relatively immobile in the temperature range considered and the covariation of $^{40}\text{Ar}/^{38}\text{Ar}$ with Ar retention (Fig. 4b) mainly reflects diffusion of ^{40}Ar from several different low-temperature sites (JESSBERGER *et al.*, in preparation). The correlation is significant at >99% confidence level ($r = 0.81$, $N = 9$) and, again, the trend cannot be explained by simple mass fractionation since, as in the $^{21}\text{Ne}/^{22}\text{Ne}$ case, the overall trend is opposite in direction and more extreme than that predicted by mass fractionation.

Apparent activation energies

As in Papers I–VI, we assume that the rate-determining step for mobile element loss during heating

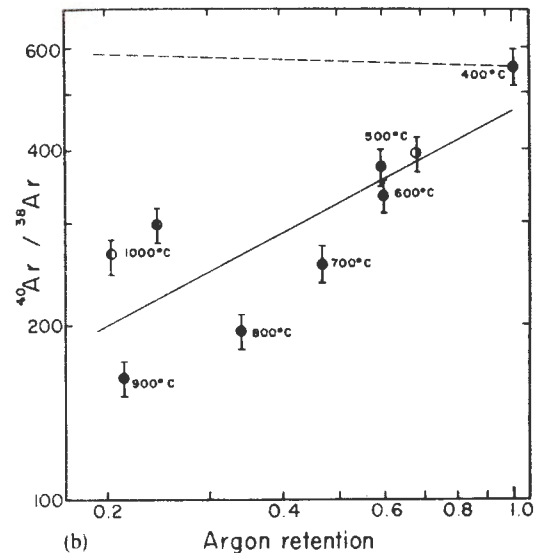
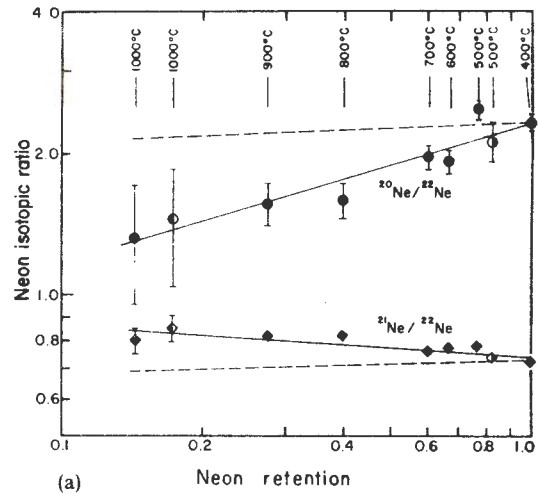


Fig. 4. $^{20}\text{Ne}/^{22}\text{Ne}$, $^{21}\text{Ne}/^{22}\text{Ne}$ and $^{40}\text{Ar}/^{38}\text{Ar}$ as a function of fractional elemental retention (which, in turn, varies with temperature). Half-filled symbols designate Ne isotopic ratios (Fig. 4a) or $^{40}\text{Ar}/^{38}\text{Ar}$ (Fig. 4b) in samples heated under non-standardized conditions. All three isotopic ratios are more variable than should occur by mass-fractionation (dashed lines) and trends of $^{21}\text{Ne}/^{22}\text{Ne}$ and $^{40}\text{Ar}/^{38}\text{Ar}$ are of opposite direction indicating that variations reflect differential outgassing of specific reservoirs with characteristic isotopic composition.

can be treated as occurring via diffusion from spherical grains of uniform size such that a diffusion parameter, D/a^2 , can be calculated from the relative retention, C/C_0 , after isothermal heating [cf. eqn (1) of Paper I]. For data obeying the Arrhenius equation [cf. eqn (3) of Paper I], the relationship between $\log D/a^2$ and reciprocal absolute temperature defines a straight line whose slope corresponds to the apparent activation energy, Q , for the loss process. FECHTIG *et al.* (1963) calculated Q values for several noble gas nuclides in some meteorites using a similar approach but with data obtained by step-wise heating.

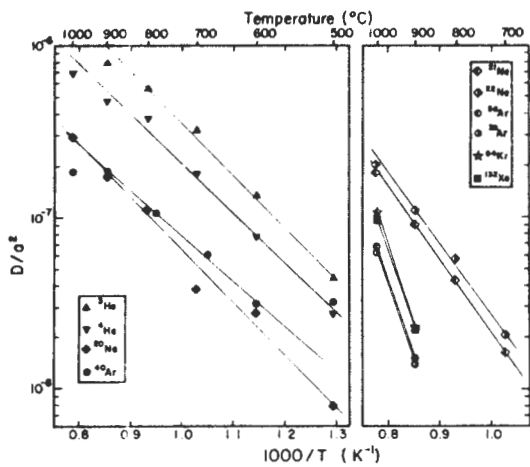


Fig. 5. Arrhenius diagrams for noble gas isotopes in heated Allende. All data are consistent with diffusive loss of each nuclide by a single process.

Arrhenius diagrams for the noble gases are illustrated in Fig. 5 and the corresponding Q values for these isotopes and carbon in Allende are listed in Table 3. [It should be emphasized that apparent activation energies calculated for ^{36}Ar , ^{38}Ar , ^{84}Kr and ^{132}Xe are based upon but two points each.] In this table we include Q values for Ne and Ar isotopes obtained from results of step-wise heating experiments on Allende (MANUEL *et al.*, 1972; SRINIVASAN *et al.*, 1978) treated as having been derived from integral release measurements. We omit Q values that could be calculated for ^{84}Kr and ^{132}Xe from step-wise heating data by these authors because, in each case, the D/a^2 values for these nuclides fail to define obvious straight lines.

In contrast with trends for several mobile elements in Allende, e.g. Bi or Tl (BART *et al.*, 1978 and in prep.), most noble gas isotopes (and carbon) define single trend lines on Arrhenius diagrams (Fig. 5). We find this somewhat surprising since each noble gas isotope would be expected to reside in various host sites with the isotopic composition for a given noble gas differing from site to site. While ^3He and ^4He may be exceptional in that data at 900 and 1000°C lie below lines extrapolated from lower temperatures, correlation coefficients of 0.99 are generated assuming that all data in the 500–1000°C range can be described by a single line. Thus there is no *a priori* statistical compulsion to assume two trend lines in either case.

One might suppose calculation of apparent activation energies for noble gases to be Procrustean in view of their known multiple host sites and reservoirs. However, even if one adopts this view, such values serve the useful purpose of facilitating mobility comparisons for they describe observed trends over a wide temperature range. Furthermore the fact that the data can be fit so readily to such a treatment argues for the validity of the approach and that the data describe loss processes rather than host-site behavior.

In general, Q values determined from our measurements of noble gas isotopes can be ordered as $\text{He} < \text{Ne} < \text{Ar} < \text{Kr} < ^{132}\text{Xe}$ (Table 3). To the extent that comparison is possible, Q values for Ne and Ar calculated from step-wise heating experiments of MANUEL *et al.* (1972) and SRINIVASAN *et al.* (1978) show similar trends. (The fit of the recalculated literature data is, as expected, poorer.) The activation energies of ^{40}Ar and, to a lesser degree, ^{20}Ne stand as exceptions to the trend above. To the extent that a single activation energy for ^{40}Ar is meaningful, its low value doubtless reflects its location in sites damaged during the decay of its parent, ^{40}K (cf. ZÄHRINGER and GENTNER, 1961). The apparent activation energies of C and the other rare gas isotopes greatly exceed values of 2–3 kcal/mole calculated for loss of Bi and Tl at 600–1000°C from Allende samples heated in the same runs that provided samples for noble gas and carbon measurements (Paper I, cf. BART *et al.*, 1978). Thus under the conditions of our experiments, we must conclude that whatever the loss mechanism, Bi and Tl are apparently more mobile than noble gas isotopes and carbon; as discussed above, retentivities of these species lead to a similar conclusion.

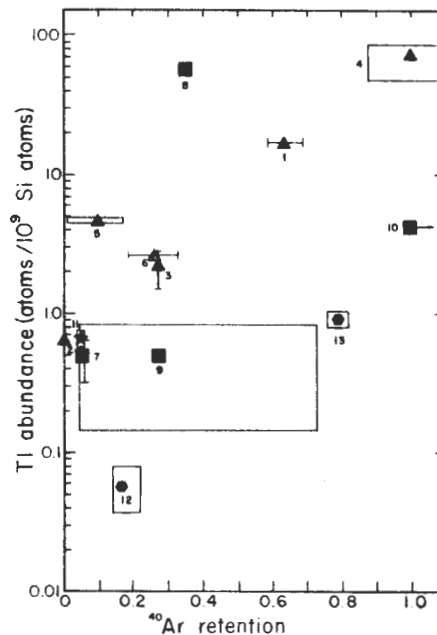


Fig. 6. Tl abundance in L-group chondrites as a function of ^{40}Ar retention. Petrologic types 3–6 are denoted by triangles, squares, stars and hexagons, respectively. Chondrites are denoted by numbers (finds in italics): (1) Bishunpur; (2) Carraweena (3) Khohar; (4) Krymka; (5) Mezö-Madaras; (6) Manych; (7) Barratta; (8) Cynthiana; (9) Goodland; (10) Tennesilm; (11) Farmington; (12) Bruderheim; (13) Modoc. Where replicate analysis exist for Tl, ^{40}Ar and/or K, we plot both the mean values and the variation resulting from the extremes among the analyses. Despite obscuration anticipated to arise from sample inhomogeneity, Tl and ^{40}Ar correlate significantly in the L-group but not in the H- or LL-groups (see text).

Gas loss by impact-heating

If elemental mobilities in the laboratory parallel those in the natural environment, as we have suggested in arguing for metamorphic alteration of E3-6 chondrites (Paper II), and if mobility trends in C3 and ordinary chondrites are similar, thermal events will affect chondritic noble gas and other mobile element concentrations. Any pronounced, open-system thermal episode should cause loss of the most mobile species present, e.g. ^3He , ^4He , ^{20}Ne and ^{40}Ar in the case of the noble gases. An early metamorphic episode would cause loss of some or all of the primordial complement but not necessarily of cosmogenic or radiogenic gas since these might not yet have been produced (but see SCHULTZ and SIGNER, 1977). Late reheating should also cause loss of all mobile species but the actual proportion lost could not be determined without knowledge of the species' initial concentrations. Of the most mobile gases, ^3He , ^4He and ^{20}Ne can be primordial and/or cosmogenic. While the proportions of each can be determined in an 'as-received' chondrite, their contents in initially unheated material can be only roughly approximated at best. ^{40}Ar , on the other hand, is very largely radiogenic and its content in matter can be estimated reasonably well if the K content and age are known.

Under the assumptions stated in the previous paragraph one might well expect a relationship between a highly-mobile trace element, e.g. Tl, and the proportion of ^{40}Ar retained in samples heated in the natural environment late in their histories after ^{40}Ar accumulation. For example, if a chondrite were shock-heated 4.55 AE after formation to an intensity sufficient to drive off both ^{40}Ar and Tl, one would expect it to have both a small amount of radiogenic ^{40}Ar and a low Tl content. An earlier shock event of equal intensity would drive off an equal amount of Tl but only that proportion of ^{40}Ar accumulated to that time. In that case a sample with a low Tl content might yet retain a substantial proportion of its radiogenic ^{40}Ar .

Among chondrites, the L-group would be expected to show best the relationship discussed above since, as a class, it apparently experienced a greater degree of late shock-heating than any other chondritic group (ANDERS, 1964; HEYMANN, 1967). To search for such an effect we culled the literature for ^{40}Ar and K data on ordinary chondrites with known Tl contents. (H- and LI-group chondrites are included as controls. These might be expected to contain a few members shock-heated at various times since formation and thus have low Tl contents not necessarily correlated with low radiogenic ^{40}Ar contents.) Published data exist for ^{40}Ar , K and Tl in 10 H-, 13 L- and 9 LL-group chondrites. However, in no case were all three species measured in aliquots of the same material and in only five instances were ^{40}Ar and K so measured. In many cases replicate data exist for a species in a given chondrite; quite often these vary considerably. (To conserve space we do not list references here;

these may be obtained from G.F.H. or M.E.L.) All of these factors should tend to cloud existing relationships, not simulate them.

Figure 6 illustrates the results for L-group chondrites. Where replicate analyses exist, we indicate the mean and overall uncertainty resulting from variation of Tl, ^{40}Ar and/or K. We estimated ^{40}Ar retention by dividing the observed ^{40}Ar content (assumed to be entirely radiogenic) by the amount that would be produced in 4.55 AE from ^{40}K decay (cf. STEIGER and JÄGER, 1977), $8.05 \times 10^{-8} \text{ cm}^3$ (STP) $^{40}\text{Ar}/\mu\text{g K}$, given the K content of the meteorite. Despite the sources of uncertainty, the data correlate at the 92% confidence level ($r = 0.51$) for the L-group; for the H- and LL- groups the data correlate at only the 41 and 55% confidence levels, respectively. In the case of the L-group, it would be desirable to minimize even the existing small probability of a chance correlation between Tl content and ^{40}Ar retention. This can only be done by determining mobile elements, K and ^{40}Ar in aliquots of the same chondritic specimens with very different gas-retention ages and we plan such an investigation in the near future.

In considering the ordinary chondrite data we have assumed that Tl abundances and ^{40}Ar retention in the L-group both reflect the same process—loss of volatiles from shock-heated parent material during post-shock cooling, estimated to range from hours (TAYLOR and HEYMANN, 1971) or weeks (SMITH and GOLDSTEIN, 1977) to years. However, an alternative, albeit less likely possibility could produce a similar trend. As a result of nebular cooling and concomitant condensation and accretion of solids by an equilibrium or quasi-equilibrium process (e.g. BLANDER and ABDEL-GAWAD, 1969; KEAYS *et al.*, 1971; WAI and WASSON, 1977) one would qualitatively expect an L-group parent object to reflect compositionally the thermal history of its parent material. Thus, material deepest in a parent body would form L6-like material at the highest temperature and contain the lowest proportion of volatile elements while material nearer the surface would form at lower temperatures and contain higher abundances of such elements (e.g. LARIMER and ANDERS, 1967; KEAYS *et al.*, 1971). Such a model explicitly assumes fractionation of volatiles during the cooling, condensation and/or accretion processes and implicitly that any alteration equivalent to or more substantial than metamorphism would occur under completely closed conditions for the elements considered. During collision, shock intensity—hence residual temperature—increases toward the interior of projectiles of decameter size (HEYMANN *et al.*, 1966; HERZOG *et al.*, 1976); whether this would occur during catastrophic break-up of objects of asteroidal size is unknown. Assuming that it would, the deepest material—containing the smallest amounts of volatiles such as Tl—would be the most heavily shock-heated and outgassed during subsequent extended cooling. If cooling occurred under conditions such that ^{40}Ar was lost but volatiles like

Tl were retained, ^{40}Ar retention would still correlate with Tl (Fig. 5). For example, as a result of prior, *in situ* shock-loading, the relative facility with which volatiles are lost in space might be altered from that observed in the laboratory. One conceivable scenario would involve the creation by outgassing of local temporary 'atmospheres' in tiny domains followed by condensation of elements like Tl during cooling of shock-heated material to low temperatures. ^{40}Ar , remaining gaseous, might then be lost relatively more easily than is observed in laboratory simulations. While we cannot ignore this possibility or other, equally speculative ones, this is not the place to discuss such ideas since they require additional experimental data. The large number of assumptions required, coupled with the fact that the apparent activation energy for loss of ^{40}Ar exceeds that of Tl in the temperature range experienced by many L-group chondrites (TAYLOR and HEYMANN, 1971; SMITH and GOLDSTEIN, 1977; cf. MATZA and LIPSCHUTZ, 1978) renders such processes less attractive alternatives.

CONCLUSIONS

Our experiments demonstrate that, as for other mobile elements, losses of C and S and noble gas isotopes progress with temperature during week-long heating of Allende in a low pressure environment. Loss of S is surprisingly small and generally parallels that of Se indicating the portions mobile at $\leq 1000^\circ\text{C}$ are sited similarly. Carbon loss, while more substantial, is less severe than anticipated as are losses of noble gas isotopes. Generally, losses are $\text{He} > \text{Ne} \sim \text{Ar} > \text{Kr} \sim \text{Xe}$ but specific isotopes vary substantially from this trend. The variation does not reflect mass-fractionation but rather preferential loss from different reservoirs. In particular, trapped Ne is not retained as well as cosmogenic Ne and radiogenic ^{40}Ar is clearly more mobile than trapped and/or cosmogenic Ar. Losses of Bi, Tl and In surprisingly exceed those of most noble gas isotopes in Allende samples.

Data for C and noble gas isotopes can each be well fit to a single straight line on Arrhenius diagrams and the magnitude of the apparent activation energies for their loss are generally consistent with diffusion-controlled processes. This suggests that the bifurcate lines observed for Bi and Tl in Allende samples treated similarly, result from differences in loss mechanism rather than from emptying of different host sites. Apparent activation energies for C and noble gas loss surprisingly exceed those for Bi and Tl from $600\text{--}1000^\circ\text{C}$ suggesting that Bi and Tl are unusually mobile. In fact, a search of the literature hints strongly that at least one of these could have been mobilized from shock-heated L-group chondrites during extended cooling although it is possible to interpret the data in a less straightforward manner. As is usual, these results answer some questions but pose other intriguing ones.

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