26 December 1975, Volume 190, Number 4221 **SCIENCE**

Host Phase of a Strange Xenon Component in Allende

Most of the noble gases reside in three rare minerals comprising less than 0.5 percent of the meteorite.

Roy S. Lewis, B. Srinivasan, Edward Anders

One of the unsolved problems of cosmochemistry is the origin of a peculiar xenon component in primitive meteorites, first discovered 11 years ago by Reynolds and Turner (*1*) in the C2 carbonaceous chondrite Renazzo. Like other primitive meteorites, Renazzo contains large amounts of primordial noble gases trapped from the solar nebula. When Reynolds and Turner heated the meteorite from 200° to 1600°C in 100°, 1-hour steps, they found that the xenon in the 700° to 1000°C fractions was enriched in the heavy isotopes 131, 132, 134, and 136 by up to 6 percent, compared to the remaining fractions. Similar or even greater enrichments were later found in other carbonaceous chondrites (*2–5*). Figure 1 shows an example from the present study.

It seemed at first that spontaneous fission of a heavy element was the most likely source of this xenon component (*1–3*). The isotopic pattern, as confirmed by several other authors (*3, 6*), somewhat resembled that of xenon from spontaneous fission of uranium-238 or other actinides, with its characteristic rise from mass 131 to 136. But the match failed in detail. None of the known actinides of reasonable half-life ($> 10^3$ years) and substantial fission decay branch showed so steep a rise from 132 to 136. This point is illustrated in Fig. 2, which compares three typical actinide spectra (*7*) with the meteoritic spectrum determined in this work.

Superheavy element? A speculative solution to this problem was proposed almost simultaneously by three groups of authors: Srinivasan *et al.* (*8*), Anders and Heymann (*9*), and Dakowski (*10*). Noting that theoretical calculations predicted an "island of stability" (better "longevity") near proton number 114 and neutron number 184 (*11*), they suggested that a superheavy element from this region might be the progenitor of the fission-derived Xe in primitive meteorites.

Anders and Heymann noted, moreover, that the anomalous Xe component correlated with volatile elements such as Hg, In, Tl, or Bi, but not with actinides such as U and Th. This suggested that the progenitor itself was volatile, and focused suspicion on elements 112 to 119, the only transuranium elements expected to be volatile. In a later paper, Anders and Larimer (*12*) narrowed the range to 111 to 116, by comparing the estimated heat of vaporization of the putative superheavy element with values extrapolated from the periodic table.

Other models. The idea of a superheavy element has not been universally accepted, and some authors even dispense with a radioactive progenitor altogether. Kuroda, Manuel, and co-workers (*4, 5*) suggested that the enrichment of the heavy Xe isotopes was caused by diffusive mass fractionation. Later Manuel *et al.* (*13*) abandoned this suggestion, after making the important observation that the light Xe isotopes 124, 126, and 128 were enriched in the fractions richest in the heavy isotopes (a trend first noted by Reynolds and Turner but then forgotten; it is also evident in Fig. 1). They suggested that both anomalies were established in some earlier locale before accretion: either in a supernova by proton and neutron capture (*p* and *r* processes), or in the early solar system by neutron reactions and diffusive mass fractionation. According to the supernova hypothesis, further developed by Black (*14*), the anomalous Xe was trapped in solid grains that were eventually incorporated in meteorites. This possibility does not seem farfetched, because several isotopically distinct, apparently presolar components have recently been discovered in primitive meteorites, on the basis of isotopic anomalies in oxygen (*15*), magnesium, and neon (*16*).

Other authors still invoke a progenitor, but allow it to decay before reaching the solar system. Clayton (*17*) and Howard *et al.* (*18*) proposed that the Xe came from a short-lived heavy element (such as ^{250}Cm, half-life 1.13×10^4 years) freshly synthesized in a supernova and condensed in dust grains in the expanding envelope. By the time these grains reached the solar system, the element had completely decayed, and so the correlation of fission Xe with volatiles would reflect the volatility of Xe, not of its progenitor.

Experimental Approaches

All of the studies cited above had been done on bulk meteorites, with the gases extracted in one or several steps. It was obvious that not much further progress could be made until the host phase of the anomalous Xe component had been isolated (*9, 12*). Our first three attempts in this direction failed. No significant enrichment of anomalous Xe was found in the two major minerals of the Orgueil C1 chondrite, a hydrated iron-magnesium silicate and magnetite (*19*), or in density fractions of

---

Roy S. Lewis and B. Srinivasan are research associates and Edward Anders is Horace B. Horton professor at the Enrico Fermi Institute and Department of Chemistry, University of Chicago, Chicago, Illinois 60637.

the silicate (*20*). A search for fission tracks in chondrules or olivine crystals from the Allende C3 and Murchison C2 chondrites likewise gave negative results (*21*), but these were inconclusive because of the low thermal stability of tracks in these minerals.

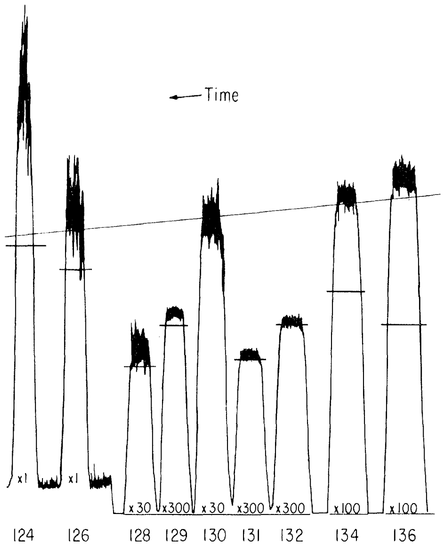

Fig. 1. Mass spectrum of Xe from Allende mineral fraction 3CS4. Heaviest and lightest isotopes are strongly enriched over their abundance in trapped meteoritic Xe (horizontal bars, normalized at the shielded isotope ^{130}Xe). The excess at masses 134 and 136 may be due to an extinct heavy element that decayed by spontaneous fission. The slanted line passing through ^{130}Xe shows the effect of spectrometer pumping as a function of time.

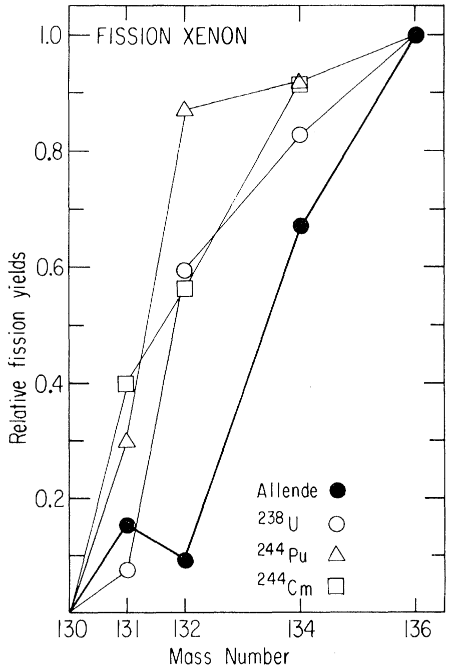

Fig. 2. Fission spectrum of unknown element from Allende meteorite, compared with spectra for three actinides (*7*) that decay by spontaneous fission. Errors are smaller than the point symbols. No known actinide matches the steep rise of the meteoritic spectrum from 131 to 136.

Our fourth attempt has been successful. This article describes the isolation and characterization of the host phase from the Allende C3V chondrite and a mass spectrometric study of the five noble gases in this phase (*22*). These studies shed new light on the origin of both the primordial noble gases in meteorites and the anomalous Xe. Although some important questions remain, we shall interpret this work as supporting a fission origin of the anomalous heavy Xe. The companion article by Anders *et al.* (*23*) describes attempts to characterize the putative progenitor of the fission Xe, by neutron activation analysis of the host phase for 26 congeners of actinides and superheavy elements.

Search for Host Phase

The only available clue to the nature of the host phase was the peak release temperature of the anomalous Xe component, 800° to 900°C. It focused suspicion on four minerals from Allende, each comprising between 1 and 4 percent of the meteorite (*24*): the sulfides troilite (FeS) and pentlandite [$(Fe,Ni)_9S_8$], and the nickel-iron alloys kamacite (~6 percent Ni) and awaruite (~68 percent Ni). Although all except pentlandite have melting points well above 1000°C, they form eutectics with each other in the range 900° to 1000°C. A further argument favoring these minerals was that volatile superheavy elements of atomic number 111 to 116 are expected to condense from the solar nebula as metals or sulfides (*12*).

We therefore prepared nickel-iron and sulfide concentrates from the Allende meteorite, by magnetic and flotation methods (*25*). Because pentlandite, in contrast to troilite and nickel-iron, was reported to be insoluble in dilute hydrochloric acid, we also attempted to prepare a pentlandite concentrate by treating the bulk meteorite alternately with HCl and HF. As it turned out, the argument relating release temperature to melting point and the alleged insolubility of pentlandite both were in error, but these two false leads nonetheless led to the right answer.

Both the metal and sulfide concentrates 1M and 1SU contained only minor amounts of anomalous Xe (Table 1) (*26*). Although the ^{136}Xe/^{132}Xe ratios resembled that of the bulk meteorite (all were higher than the primordial ratio of 0.310), the absolute amounts of Xe, as given by the ^{132}Xe contents, were markedly lower. In contrast, the acid-resistant residue 1CS was enriched in ^{132}Xe by a factor of 54, and since its ^{136}Xe/^{132}Xe ratio was similar to the ratio for bulk Allende, it must have been enriched in anomalous Xe by about the same factor. All samples showed the characteristic rise in 136/132 ratio at 1000°C.

It seems striking that a fraction com-

Table 1. Xenon in mineral fractions from Allende (*25*). Symbols: M, metal-enriched; SU, sulfide-enriched; CS, chromite, spinel, and elemental C and S. Sample sizes are given in parentheses. Gases were extracted by stepwise heating in 15-minute steps at the temperatures indicated (*26*); such temperature fractions are given in italics. Complete data are available from the authors on request. Gas content is accurate to ± 20 percent.

Sample	^{132}Xe ($\times 10^{-10}$ cm^3 STP/g)	^{129}Xe/^{132}Xe	^{136}Xe/^{132}Xe
Bulk	19.2	1.764 ±.003	0.3353 ±.0004
1M 500°C	*0.38*	*1.087* *±.015*	*0.3259* *±.0024*
1000°C	*1.33*	*1.927* *±.021*	*0.3674* *±.0028*
1600°C	*3.43*	*1.662* *±.018*	*0.3192* *±.0021*
1M total (161.7 mg)	5.14	1.688 ±.013	0.3322 ±.0016
1SU 700°C	*0.66*	*1.215* *±.022*	*0.3285* *±.0057*
1000°C	*0.97*	*3.101* *±.063*	*0.3656* *±.0073*
1600°C	*12.7*	*1.294* *±.015*	*0.3308* *±.0025*
1SU total (9.9 mg)	14.3	1.413 ±.0014	0.3330 ±.0023
1CS 500°C	*50.3*	*1.988* *±.028*	*0.3164* *±.0022*
1000°C	*173*	*1.386* *±.015*	*0.3719* *±.0023*
1600°C	*817*	*1.046* *±.012*	*0.3282* *±.0019*
1CS total (18.9 mg)	1040	1.148 ±.010	0.3349 ±.0015

Table 2. Electron microprobe analyses of Allende mineral fractions. Compositions were measured on compacted powders by averaging the beam trace over a typical area. Because of sample heterogeneity and poor geometry, relative errors seem to have been on the order of 20 to 50 percent, as shown by the differences between the two 4CS1 analyses and between the microprobe and neutron activation values for Cr and Fe. In calculating the sums appropriate amounts of oxygen were added.

Element	Content (weight percent)		
	4C1	4CS1	4CS1
Mg	0.08	2.3	3.1
Al	0.87	6.62	8.4
Si	0.21	0.12	0.17
S	1.33	0.75	0.95
Cr	7.79 14.3*	8.43 8.0*	9.25
Fe	4.63 10.2*	5.79 8.6*	6.89
Sum (+ O)	36.9	39.7	44.2
"Carbon"†	~ 60		

*Instrumental neutron activation analysis results of Gros (*30*). †Weight loss on treatment with atomic oxygen.

prising only 1 percent of the meteorite should contain ∼50 percent of the primordial and anomalous Xe, although somewhat similar but less extreme cases have been reported before (*27*). Actually, the enrichment was even greater. Sample 1CS turned out to contain 50 percent free sulfur, which was subsequently removed by CS_2. X-ray diffraction and electron microprobe analysis of the residue showed that it did not consist of pentlandite at all, but of coarse-grained ($\leq$ 20 μm) spinel, $MgAl_2O_4$, and fine-grained (0.3 μm) chromite, $FeCr_2O_4$ (*28*).

It turned out that most of the gas resides in the chromite rather than the spinel. A spinel-free chromite sample was prepared by digesting 22.6 g of Allende (crushed to 2 to 5 mm) with HF and HCl until it had partially disintegrated, and decanting the fine fraction that did not settle 10 cm in 30 seconds. On further treatment with HF, HCl, and CS_2, this fraction yielded 38 mg of a fine-grained residue (3C1), which showed only chromite lines in its x-ray diffraction pattern, and no transparent spinel grains under the microscope. The coarse fraction, after similar treatment, gave a residue of 70 mg (3CS1), which contained both chromite and spinel. The spinel-free sample 3C1 had a 50 percent higher Xe content than the spinel-bearing 3CS1, which suggests that most of the Xe is contained in the chromite. This is not surprising, because the spinel comes from the Ca,Al-rich inclusions, which are high-temperature condensates (*29*) and hence should have taken up only small amounts of gas from the solar nebula. The chromite, on the other hand, occurs largely in the matrix (*24*), which is a low-temperature condensate generally rich in volatiles (*29*).

Electron microprobe analysis of two analogous samples, 4C1 and 4CS1, showed mainly Fe, Cr, S, Al, and Mg (Table 2). The analyses fell far short of adding up to 100 percent, in part because of systematic errors in the measurement of fine powders [compare the microprobe data for Cr and Fe with neutron activation values (*30*) for the same elements], but mainly because the sample contained a large amount of carbonaceous matter (about 60 percent, judging from the weight loss on treatment with atomic oxygen). Breger *et al.* (*31*) had previously found 65 percent carbon in a similar chromite concentrate from Allende, and had established that the carbon was amorphous to x-rays and free of H and N. Mössbauer spectroscopy by Virgo (*32*) showed that the chromite contained some 40 percent Fe_3O_4 in solid solution.

The samples consistently showed small amounts of sulfur (average atomic Cr/S ratio = 6). However, the x-ray diffraction pattern failed to show any lines of minerals other than spinel and chromite, although a special search was made for all known sulfides of Fe and Cr as well as the sulfospinel daubrulite, $FeCr_2S_4$. Possibly this reflects insufficient analytical sensitivity. X-ray diffraction often misses minerals with an abundance of less than 5 percent, particularly if they are very fine-grained. For example, chromium (III) sulfide prepared at temperatures as high as 450°C is amorphous to x-rays (*33*).

Etching Experiments: Mineral Q

It did not make sense for chromite to be the host phase of either primordial or the progenitor of fission-derived Xe. The cations of chromite, Fe^{2+} (0.71 Å) and Cr^{3+} (0.70 Å), are too small to permit substitution by the large positive ions of actinides or superheavy elements, and the anion O^{2-} (1.32 Å) likewise is too small for ready replacement by neutral Xe (1.90 Å). The situation for amorphous carbon was hard to predict a priori, because of its irregular structure. On the other hand, a sulfide (S^{2-}, 1.72 Å) would be a much more appropriate host phase for Xe, as pointed out by DuFresne and Anders (*34*). Moreover, the superheavy elements 111 to 116 were expected to condense as metals or sulfides, not as oxides (*12*).

Apart from these theoretical reasons, there were two experimental hints of the presence of a minor sulfide mineral: the occurrence of S in the electron microprobe analysis, and the high abundance of Se, Te, Bi, and other sulfide-seeking trace elements in sample 3CS1 (*23*). Such a mineral might be present as discrete grains, or as a surface coating on chromite.

It is always difficult to isolate an unknown mineral, and impossible if it forms a surface coating on another mineral. For this reason we chose a different approach:

Table 3. Helium, neon, and argon from acid-treated mineral fractions of the Allende meteorite. Symbols: 3C, chromite; 3CS, chromite-spinel; 1, unetched; 2, HNO_3 etch; 3, aqua regia etch; 4, HNO_3, $HClO_4$ etch; HB, hot blank; CB, cold blank. All samples were heated to 1600°C for 30 minutes to extract noble gases, except 3CS2, which was analyzed in four steps at the temperatures indicated. Unless otherwise stated, abundances are accurate to ± 20 percent. Isotopic ratios have been corrected for mass discrimination, background of the spectrometer, and hot blank. One-half of the latter two corrections was included in the error.

Sample	^{4}He (× 10^{-8} cm^3 STP/g)	^{22}Ne (× 10^{-8} cm^3 STP/g)	^{20}Ne/^{22}Ne	^{21}Ne/^{22}Ne	^{36}Ar (× 10^{-8} cm^3 STP/g)	^{36}Ar/^{38}Ar	^{40}Ar/^{36}Ar
Bulk	2,550§	2.50	2.71 ±.03	0.713 ±.004	16.7§	5.25§	89.2§
3C1 (3.98 mg)	189,000	115	9.44 ±.15	0.04231 ±.00075	3,715	5.255 ±.034	3.95 ±.08
3C3 (3.33 mg)	195,000	118	9.24 ±.21	0.0415 ±.0010	1,575	5.230 ±.042	2.00 ±.22
3C2 (2.93 mg)	169,000	111	8.53 ±.17	0.04346 ±.00082	390	5.213 ±.074	1.1 ±1.0
3CS1 (4.32 mg)	139,000	81	8.64 ±.16	0.05536 ±.00092	2,430	5.270 ±.034	3.89 ±.11
3CS3 (4.69 mg)	109,000	67	8.53 ±.17	0.0542 ±.0010	1,025	5.265 ±.038	1.71 ±.25
*3CS2 700°C**	*8,200* *±4,200*	*0.4* *±1.4*	*11* *±44*	*0.4* *±1.3*	*45*	*5.25* *±.22*	*0*
*1000°C**	*76,800*	*34*	*8.46* *±.36*	*0.0452* *±.0017*	*39*	*4.92* *±.25*	*0*
*1300°C**	*45,600*	*41*	*8.25* *±.29*	*0.0602* *±.0018*	*143*	*5.023* *±.076*	*0*
*1600°C**	*1,500* *±4,000*	*0.4* *±1.1*	*5* *±25*	*0.3* *±1.1*	*11*	*5.18* *±.97*	*0*
3CS2 Total* (3.54 mg)	122,400†	75†	8.35† ±.23	0.0534† ±.0013	238	5.054 ±.084	0
3CS4‡ (1.41 mg)	163,000	95	8.45 ±.36	0.0558 ±.0022	185	4.97 ±.14	0
HB 1600°C (30 min)	30	6 × 10^{-3}	∼7.1	∼0.03	8 × 10^{-3}	∼5.3	∼295
CB 25°C (30 min)	30	2 × 10^{-3}	∼7.1	∼0.03	7 × 10^{-5}	∼5.3	∼295

*Corrected for hot blank since no blanks were measured at intermediate temperatures. However, if cold blank corrections are applied, the reported isotopic ratios and abundances remain unchanged within the limits of experimental error, except for the ^{40}Ar/^{36}Ar ratios, which become 11.07 ± 0.10 at 700°C, 1.07 ± 0.08 at 1000°C, 0.192 ± 0.021 at 1300°C, and 2.426 ± 0.027 for total. †The summation excludes the 700° and 1600°C fractions for He and Ne. ‡The He and Ne contents of this very small sample are higher than those of the remaining samples, in spite of the more severe etching treatment. This suggests either a weighing error or loss of a gas-poor phase on etching. For comparison with other samples, the gas contents of 3CS4 should therefore be multiplied by 0.68. The isotopic ratios remain unchanged, of course. § (*36*).

selective dissolution in an appropriate solvent (*19, 35*). All sulfides are soluble in hot nitric acid or aqua regia, although some [including several chromium sulfides (*33*)] react only slowly. Chromite and carbon, on the other hand, are insoluble in these acids. We therefore treated portions of samples 3C and 3CS with acids for varying lengths of time, as shown below, and analyzed them by mass spectrometry and neutron activation analysis (*23*).

3C2, 3CS2	Concentrated HNO_3, 3.5 hours, 80°C
3C3, 3CS3	Aqua regia, 1 hour, 40°–80°C; CS_2; aqua regia, 2.5 hours, 40°–80°C
3CS4	Red fuming HNO_3, 1 hour, at 80°C; concentrated $HClO_4$, 1 hour, 80°C

Table 4. Krypton from acid-treated mineral fractions of the Allende meteorite (*26*). Symbols are defined as in Table 3. Gas concentrations are accurate to ± 20 percent.

Sample	^{84}Kr (×10^{-10} cm^3 STP/g)	^{78}Kr*	^{80}Kr	^{82}Kr	^{83}Kr	^{86}Kr
		^{84}Kr ≡ 100				
Bulk	26.2	0.613	5.268	20.621	20.187	30.96
		±.039	±.015	±.039	±.048	±.033
3C1	3827	0.6030	3.969	20.153	20.209	31.043
(3.98 mg)		±.0060	±.037	±.097	±.085	±.082
3C3	1672	0.6074	3.921	19.99	20.11	31.190
(3.33 mg)		±.0088	±.040	±.12	±.11	±.095
3C2	389	0.570	3.760	19.08	20.024	32.067
(2.93 mg)		±.033	±.063	±.15	±.099	±.095
3CS1	2361	0.6085	4.009	20.256	20.270	31.121
(4.32 mg)		±.0076	±.045	±.091	±.078	±.083
3CS3	1092	0.5967	3.928	19.969	20.115	31.200
(4.69 mg)		±.0098	±.043	±.097	±.096	±.092
_3CS2 700°C_	_65_	_0.61_	_4.052*_	_20.52_	_20.66_	_30.75_
		_±.14_	_±.070_	_±.21_	_±.16_	_±.15_
_1000°C_	_32_	_0.62_	_3.583*_	_18.66_	_19.91_	_33.82_
		_±.25_	_±.078_	_±.28_	_±.26_	_±.25_
_1300°C_	_118_	_0.534_	_3.649*_	_18.33_	_19.98_	_32.98_
		_±.066_	_±.046_	_±.17_	_±.14_	_±.10_
_1600°C_	_13_	_0.81_	_3.85*_	_18.69_	_19.51_	_32.35_
		_±.59_	_±.10_	_±.31_	_±.30_	_±.38_
3CS2 total	228	0.58	3.766*	19.08	20.160	32.38
(3.54 mg)		±.07	±.033	±.11	±.090	±.11
3CS4	147†	0.51	3.359*	17.69	19.44	33.73
(1.41 mg)		±.15	±.044	±.23	±.23	±.17

*Corrections for hydrocarbon background in the mass spectrometer have been applied to all ^{78}Kr peaks and some ^{80}Kr peaks. Half of the correction was included in the error. †For comparison with other samples, the ^{84}Kr content of 3CS4 should be multiplied by 0.68 [see third footnote (‡) in Table 3].

Table 5. Xenon from acid-treated mineral fractions of the Allende meteorite (*26*). Symbols are defined as in Table 3; $^{136}Xe_f$ = fission-derived ^{136}Xe. Gas concentrations are accurate to ± 20 percent.

Sample	^{132}Xe ($^{136}Xe_f$) (× 10^{-10} cm^3 STP/g)	^{124}Xe	^{126}Xe	^{128}Xe	^{129}Xe	^{130}Xe	^{131}Xe	^{134}Xe	^{136}Xe
		^{132}Xe ≡ 100							
Bulk	19.2	0.4629	0.4019	8.239	176.4	16.04	81.71	39.16	33.53
	(0.50)	±.0034	±.0038	±.018	±.3	±.03	±.10	±.05	±.04
3C1	3033	0.4712	0.4109	8.152	113.88	16.09	81.83	39.31	33.83
(3.98 mg)	(88)	±.0068	±.0044	±.043	±.76	±.12	±.66	±.20	±.21
3C3	1402	0.4911	0.4233	8.341	113.84	16.16	82.09	40.32	35.44
(3.33 mg)	(65)	±.0084	±.0043	±.037	±.79	±.11	±.58	±.21	±.22
3C2	421	0.586	0.4565	8.550	110.33	15.92	82.94	47.25	46.11
(2.93 mg)	(66)	±.013	±.0094	±.067	±.77	±.12	±.62	±.30	±.31
3CS1	1984	0.4735	0.4185	8.187	114.25	16.16	81.87	39.23	33.81
(4.32 mg)	(60)	±.0075	±.0044	±.036	±.74	±.12	±.58	±.20	±.22
3CS3	970	0.4922	0.4243	8.377	112.23	16.31	82.78	40.42	35.40
(4.69 mg)	(42)	±.0071	±.0067	±.061	±.97	±.16	±.68	±.23	±.25
_3CS2 700°C_	_55_	_0.420_	_0.386_	_8.172_	_124.42_	_16.14_	_81.32_	_38.80_	_32.61_
		_±.013_	_±.018_	_±.071_	_±.94_	_±.13_	_±.70_	_±.31_	_±.24_
_1000°C_	_36_	_0.688_	_0.494_	_8.841_	_115.02_	_15.74_	_83.06_	_52.73_	_54.50_
		_±.011_	_±.010_	_±.094_	_±.92_	_±.15_	_±.69_	_±.35_	_±.45_
_1300°C_	_152_	_0.668_	_0.510_	_8.783_	_105.4_	_15.94_	_84.11_	_50.80_	_51.60_
		_±.014_	_±.012_	_±.076_	_±1.1_	_±.16_	_±.68_	_±.27_	_±.34_
_1600°C_	_15_	_0.630_	_0.483_	_8.59_	_106.7_	_15.97_	_82.7_	_47.62_	_46.38_
		_±.034_	_±.010_	_±.14_	_±1.4_	_±.20_	_±1.6_	_±.38_	_±.37_
3CS2 total	258	0.6159	0.4798	8.650	110.87	15.96	83.29	48.33	47.65
(3.54 mg)	(44)	±.0091	±.0082	±.050	±.70	±.10	±.45	±.18	±.22
3CS4	198*	0.727	0.487	8.78	106.14	15.84	83.63	55.96	58.32
(1.41 mg)	(54)	±.016	±.023	±.11	±.94	±.14	±.72	±.41	±.47

*For comparison with other samples, the ^{132}Xe and $^{136}Xe_f$ contents of 3CS4 should be multiplied by 0.68 [see third footnote (‡) in Table 3].

The noble-gas data were rather striking (Tables 3 to 5). Although the acid-treated samples lost only 4 to 8 percent of their weight (*23*), they lost a major part of their heavy noble gases Ar, Kr, and Xe, retaining only about 45 percent in aqua regia, 10 percent in concentrated HNO_3, and 5 percent in fuming HNO_3. Yet the light gases He and Ne and the anomalous Xe were essentially unaffected, as shown by the constancy of the He and Ne contents and the rise in $^{136}Xe/^{132}Xe$ and $^{124}Xe/^{132}Xe$ with falling ^{132}Xe content. The concentration of $^{136}Xe_f$ is given in Table 5.

The isotopic enrichments in ^{136}Xe far exceed any observed heretofore. Bulk chondrite samples never have $^{136}Xe/^{132}Xe$ ratios higher than 0.349 (*36*), compared to a primordial ratio of 0.310 (this work), and even temperature fractions have never been known to exceed a ratio of 0.47 (*5*). A very substantial separation of trapped Xe from fission Xe has been achieved.

Apparently the heavy noble gases reside mainly in a minor mineral fraction, soluble in oxidizing acids. We have dubbed it Q, for quintessence (*37*). The exact nature of this fraction is still uncertain, but according to data presented in (*23*), it contains Fe, Cr, Ni, and Al in proportions corresponding to a M^{2+}/M^{3+} ratio between 1 and 2. It may be a single mineral (perhaps an unknown double sulfide of Fe and Cr, such as $Fe_4Cr_2S_7$ or $Fe_2Cr_2S_5$), or a mixture of several minerals. A detailed discussion is given in (*23*).

Host Phase: Q or Chromite-Carbon?

Much of the discussion of this paper will focus on the differences between Q and chromite-carbon, and we have therefore listed the gas contents of both in Table 6. For chromite-carbon, we used the most severely etched sample, 3CS4, although the trace element composition of a similar sample, 3CS5 (*23*), suggests that it still contained some Q. For the gas content of Q, we used the difference between unetched and etched samples, for example

$$[Q(C2)] = \left\{[C1] - (1 - \alpha)[C2]\right\}/\alpha \quad (1)$$

where brackets denote the gas concentration of a sample, Q(C2) is Q as inferred from sample 3C2, and α is the fractional weight loss on etching [0.075 for 3C2 and 0.064 for 3CS2 (*23*)]. As a check on the method, we calculated gas contents of Q for both the chromite and chromite-spinel series. The two estimates agree fairly well (Table 6).

Evidently, Q has exceedingly high contents of heavy noble gases, some 300 times higher than the chromite-carbon, which in turn is enriched by an order of magnitude over the bulk meteorite. The light noble gases are enriched about equally in both minerals, by nearly two orders of magnitude over the bulk meteorite.

Table 6 also brings out an exceedingly important point: fission Xe is some seven to ten times more abundant in Q than in chromite-carbon. To be sure, 3CS4 contains at least three minerals: chromite (20 to 25 percent), carbon (~60 percent), and spinel (15 to 20 percent). If only one of them contained fission Xe, the underabundance relative to Q would no longer be so striking. However, analyses of separated carbon and chromite fractions (*38*) show that fission Xe is present in both. (Therefore we shall frequently use chromite hereafter as a synonym for chromite-carbon.) Barring the possibility that fission Xe is contained in some minor mineral hidden within these fractions, it would seem that Q is indeed the phase with the highest concentration of fission Xe.

The present distribution of fission Xe may not accurately reflect the distribution of the progenitor, because of recoil of fission fragments into adjacent phases. But except for some highly artificial configurations, such recoil cannot reverse an original gradient, enriching a catcher phase over the parent phase. Thus Q must be the parent phase.

Elemental Patterns

All samples have very low ^{21}Ne/^{22}Ne and ^{40}Ar/^{36}Ar ratios relative to bulk Allende. This shows that the gases are almost entirely primordial (or trapped), with only small admixtures of cosmic-ray produced ^{21}Ne and radiogenic ^{40}Ar. From the elemental abundance patterns, it is clear that the gases are of planetary (*39*), not solar type (Fig. 3). Relative to cosmic abundances, the heavier gases are strongly enriched, as in other carbonaceous chondrites and the earth's atmosphere, but unlike solar wind in lunar soil (*40*) or gas-rich meteorites [such as Pesyanoe (*41*)], which gives a gently rising pattern (Fig. 3). We have not plotted Q itself, but it has a pattern even steeper than the conventional planetary pattern, with about 200 times as much He and Ne and about 2000 times as much Ar, Kr, and Xe as are present in bulk Allende.

Table 6. Gas contents of mineral Q and chromite-carbon. Errors are based on uncertainties in elemental abundances (except for isotopic ratios, where only the error in ratio was used). For ^{136}Xe$_f$ in 3CS4, the measured values were reduced by a factor of 0.68, as indicated in the third footnote (‡) to Table 3. Doubtful values are given in parentheses. Concentrations [Q(C2)] and [Q(CS2)] are defined as in Eq. 1.

Element or ratio	Mineral Q (× 10^{-8} cm^3 STP/g)		Chromite 3CS4 (× 10^{-8} cm^3 STP/g)
	[Q(C2)]	[Q(CS2)]	
^{4}He	440,000 ±650,000	380,000 ±570,000	111,000 ±22,000
^{20}Ne	2,800 ±3,700	1,800 ±2,900	546 ±110
^{36}Ar	45,000 ±10,000	35,000 ±7,700	126 ±25
^{84}Kr	460 ±100	340 ±75	1.0 ±.2
^{132}Xe	350 ±80	270 ±63	1.35 ±.27
^{136}Xe$_f$	3.7 ±1.2	2.5 ±.9	0.36 ±.08
$\frac{^{20}\text{Ne}}{^{22}\text{Ne}}$	(17) ±2	11 ±2	8.45 ±.36
$\frac{^{36}\text{Ar}}{^{38}\text{Ar}}$	5.26 ±.04	5.29 ±.04	4.97 ±.14
$\frac{^{136}\text{Xe}}{^{132}\text{Xe}}$	0.320 ±.003	0.319 ±.003	0.583 ±.005
$\frac{^{20}\text{Ne}}{^{36}\text{Ar}}$	0.06 ±.08	0.05 ±.08	4.3 ±1.2

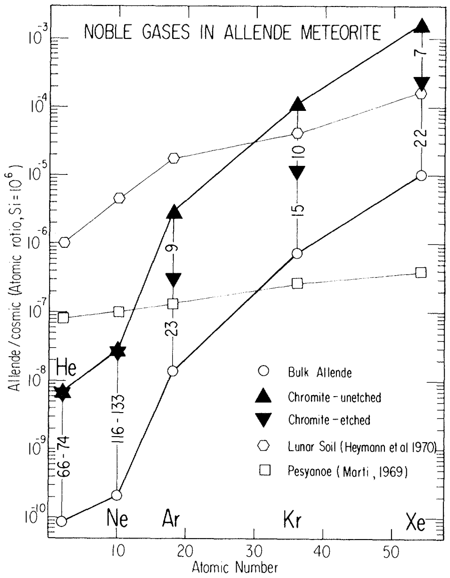

Fig. 3. Allende meteorite has a typical planetary noble gas pattern, with He and Ne showing very large but similar depletions relative to solar system abundances (*39*), while the three heavier gases show progressively smaller depletions. Such a pattern was apparently established by selective trapping of gases from the solar nebula. In contrast, solar-wind ion implantation is nonselective and gives a nearly flat pattern [Pesyanoe meteorite (*41*)], sometimes skewed by saturation and diffusion effects [lunar soil (*40*)]. The unetched chromite has nearly the same pattern as the bulk meteorite, while the etched sample shows a lower ratio of heavy to light gases. Abundances for these two samples are plotted relative to bulk meteorite. Numbers indicate the enrichment factor.

The steep patterns suggest that the gases were not trapped by ion implantation. A more selective process is implied, such as trapping in anion vacancies of a mineral forming in the solar nebula, as proposed for planetary gases in general (*34, 42, 43*).

Isotopic Patterns

There are major isotopic differences between the trapped gas in Q and chromite, which must have an important bearing on the anomalous Xe problem. Some were indicated in Table 6, others will be discussed below. To anticipate our findings: the gas in Q has essentially unfractionated isotopic composition (close to solar), despite its extremely fractionated elemental composition. The gas in chromite, on the other hand, shows substantial isotopic fractionations, varying in sign and magnitude.

So as not to degrade the isotopic data by additional corrections, we shall discuss these differences in terms of the actual samples, not the pure end members Q and chromite. This causes no real problems, because the most severely etched samples 3C2, 3CS2, and especially 3CS4 approach the pure chromite pattern, while the unetched samples 3C1 and 3CS1 are dominated by Q at least for the three heavy gases.

Neon. The Ne in primitive meteorites is a mixture of three components: solar, planetary, and cosmogenic (produced by cosmic-ray induced spallation reactions on Mg, Al, and Si). Solar Ne is dominated by the light isotope neon-20: ^{20}Ne/^{22}Ne = 12.65 and ^{21}Ne/^{22}Ne = 0.0316 (*44*). Planetary Ne (also "Neon-A") differs from solar Ne in being enriched in the heavier isotopes by about 20 percent per mass unit: ^{20}Ne/^{22}Ne = 8.2 and ^{21}Ne/^{22}Ne = 0.025. Cosmogenic Ne contains all three isotopes in about the proportions 0.85 : 0.92 : 1 (*36, 44*).

The data are shown on a conventional three-isotope plot (Fig. 4). It is a property of such a plot that binary mixtures lie on a straight line joining the two components, whereas ternary mixtures lie within a triangle whose vertices are the three components. The etched samples fall close to the planetary corner, and hence consist mainly of planetary Ne, with small amounts of cosmogenic Ne. (The latter is more abundant in the spinel-bearing samples, because Ne is produced more abundantly from Mg and Al than from Cr and Fe.) The ^{20}Ne/^{22}Ne ratio drops with increasing degree of etching, especially in the three chromite samples (triangles). Apparently they contain a less fractionated Ne component of higher ^{20}Ne/^{22}Ne ratio, of solar or intermediate composition. Because this component is selectively removed on etching, it must be associated with Q. Evidently the Ne in chromite is enriched in the heavy isotopes, while the Ne in Q is of less fractionated composition, closer to solar.

Argon. The unetched samples (whose Ar is dominated by that in Q) have ^{36}Ar/^{38}Ar ratios of 5.26 to 5.27, close to the solar or terrestrial atmospheric values of 5.33 ± 0.03 or 5.32 ± 0.01 (*45*). But the etched samples 3CS2 and 3CS4 have markedly lower ratios: 5.05 and 4.97. Again, the heavy isotope is enriched in the chromite but not in Q. There have been earlier reports of a similar enrichment in some meteorite classes (*36, 44*).

The etched samples contain little or no ^{40}Ar. The ^{40}Ar/^{36}Ar ratio in nucleosynthesis is believed to be about 2×10^{-4} (*39*), but measured ratios in chondrites are always much higher, because of a contribution from the decay of ^{40}K. The K content of the chromite was not measured, but is likely to be low, since Rb in 3C1 is less than one-third as abundant as in the bulk meteorite (*23*).

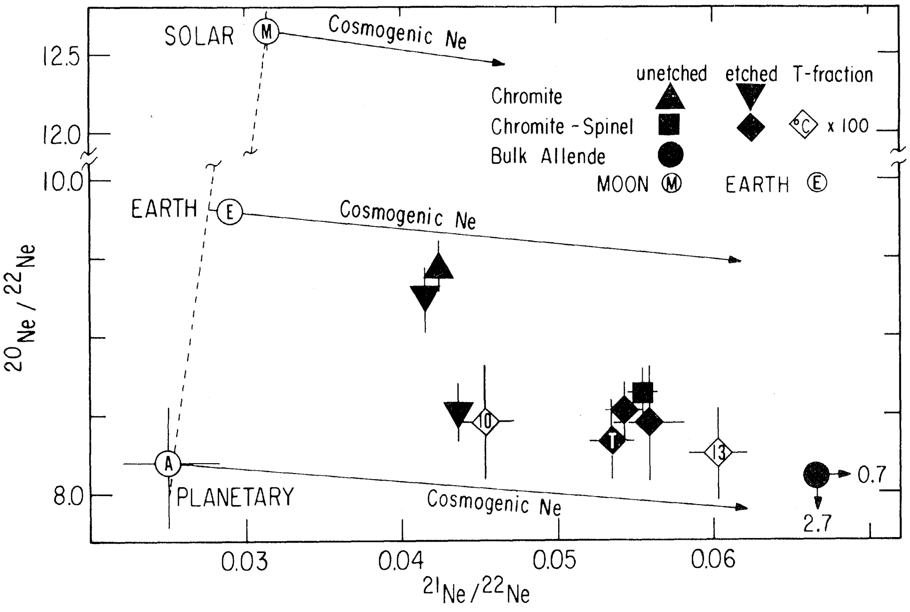

Fig. 4. Isotopic composition of Ne in Allende chromite fractions. Composition of gas is indicated by proximity to one of the three components (planetary, solar, and cosmogenic; the latter lies off scale at ^{20}Ne/^{22}Ne = 0.85, ^{21}Ne/^{22}Ne = 0.92). All samples consist mainly of planetary Ne, with small admixtures of cosmogenic Ne and a less fractionated (solar?) component. Dashed lines in this and subsequent figures indicate mass fractionation trend for solar component. Symbol T denotes total of the temperature fractions of 3CS2.

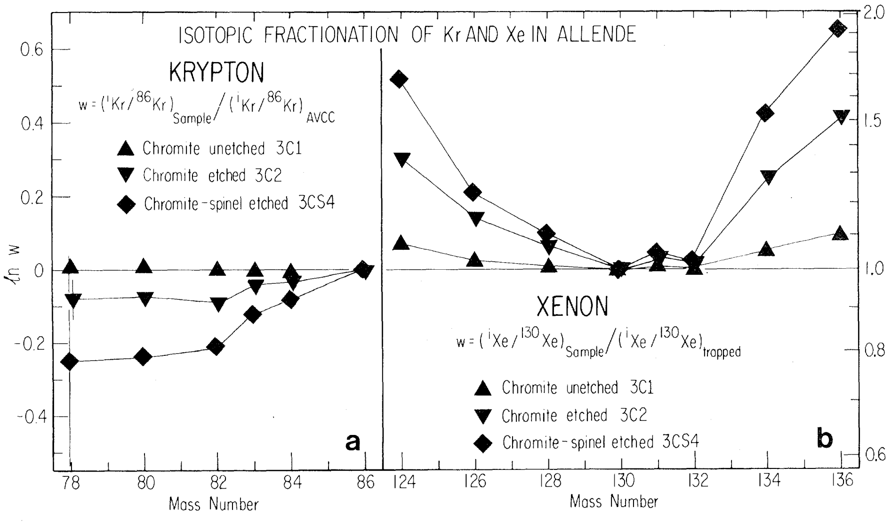

Fig. 5. (a) Krypton, like Ne and Ar, shows an enrichment of the heavy isotopes in the etched samples, relative to average carbonaceous chondrite [AVCC (*46*)] krypton. It may be due to mass fractionation, or perhaps in part to a fission contribution. (b) Xenon is more complicated, showing enrichments at both the light and heavy isotopes. The enrichment of the heavy isotopes is almost certainly due to fission, which leaves mass fractionation (but in a direction opposite to that of the other noble gases!) to explain the light isotopes.

Krypton and xenon. For these elements, with six and nine stable isotopes apiece, it is convenient to plot isotopic ratios relative to the same ratios in a reference composition (Fig. 5). The compositions chosen were average carbonaceous chondrite Kr [= AVCC Kr (*46*)] which is virtually identical to atmospheric and solar Kr, except for a 1.5 percent excess at mass 86 (*45*), and the trapped Xe component in Q, as determined in this work and reported below. The latter differs slightly from AVCC and solar Xe, but to a degree that does not affect the present analysis.

The Kr follows the trend of Ne and Ar. Again, the heavy isotopes are enriched in the etched but not the unetched samples. The isotopic pattern could be explained in terms of either mass fractionation or fission as the dominant process, but neither explanation is wholly satisfactory. Mass fractionation leaves residual variations in the isotopic ratios that are hard to explain. Fission, on the other hand, leads to improbable ^{86}Kr/^{84}Kr and ^{86}Kr/^{136}Xe yield ratios.

Mass fractionation, as invoked for Ne and Ar, should give a smooth curve (or even a straight line for the simpler kinds of fractionation). The Kr data for etched samples (Fig. 5a) do not follow this prediction, and so another mechanism must be sought.

Qualitatively, fission can account for the trend. It would enrich unshielded 83, 84, and 86, but not shielded 78, 80, and 82, in accord with observation. To test this mechanism quantitatively, we calculate the isotopic composition of the fission Kr, using three-isotope plots such as Fig. 6. On the assumption that the fission yield of shielded ^{82}Kr is zero, the x-intercept of the correlation line through the data gives the fission ^{86}Kr/^{84}Kr ratio. The fission ^{83}Kr/^{84}Kr ratio can be found from an analogous plot. In this manner we obtain the following relative yields: ^{83}Kr : ^{84}Kr : ^{86}Kr = (0.283 ± 0.023) : (2.02 ± 0.12) : 1.00. The apparent maximum at 84 is very puzzling. Fission spectra sometimes show small fine-structure peaks superimposed on a monotonic trend, but never as large as this.

To find the absolute amount of fission Kr, we need to know the composition of trapped Kr. No obvious candidate exists, but judging from the Xe data, only a small error (~10 percent) is introduced by using the unetched sample 3CS1 for the trapped component in the severely etched 3CS4. Assuming a zero fission yield of the shielded isotope ^{82}Kr, we find ^{86}Kr$_f$ = 54×10^{-10} cm^3 STP (standard temperature and pressure) per gram from sample

3CS4. Combining this result with the $^{136}Xe_f$ content derived below, we obtain $^{86}Kr_f/^{136}Xe_f \approx 0.18$. This ratio exceeds not only previous estimates of 0.06 or ~0.1 (*46*) for the chondritic fission component, but also measured values of $^{86}Kr_f/^{136}Xe_f$ for heavy actinides—for example, 0.019 for ^{244}Pu (*47*) and 0.031 for ^{252}Cf (*8*).

Other processes exist that can alter the isotopic composition of Kr (neutron capture or spallation of neighboring elements), but they would lead to observable effects that are not seen. Thus we are left with two unsatisfactory explanations for the Kr data: atypical fission or mass fractionation coupled with another, unknown process (or, of course, a combination of the two). The present data do not tell which of these processes is dominant, not even for any one isotope such as ^{86}Kr.

The Xe pattern in Fig. 5 looks strikingly different from the Kr pattern. The heavy isotopes are again enriched, but so are the light isotopes. No single process can produce such a pattern. Fission can account for the heavy isotopes and is probably the only acceptable explanation for them. But it cannot account for the four light isotopes, which are neutron-deficient and hence made only in very small yield. Nuclear reactions with charged particles could, in principle, make the light isotopes preferentially, but all known reactions of this type produce more ^{126}Xe than ^{124}Xe, and also enhance ^{78}Kr.

This leaves mass-dependent fractionation (*13*) as the remaining, if still unsatisfactory, alternative. The light isotope 124 lies above the trend line defined by the next three isotopes 126 to 130, and so some ad hoc explanation must be found for this anomaly. (Alternatively, one could assume that the fractionation line is defined by 124 and 126, but this leads to implausibly large excesses for 128 and 130.) Moreover, the sign of the fractionation is the opposite of that for the three lighter gases—the light, not the heavy isotopes are enriched. We shall return to this problem later on.

In Situ Decay?

Granted that the heavy Xe component formed by fission, did the fission take place in situ—that is, in the chromite-carbon-Q grains—or in some earlier locale? Manuel *et al.* (*13*) took the latter view, because they found no difference in the thermal release patterns of the lightest and heaviest Xe isotopes. A plot of $^{124}Xe/^{130}Xe$ against $^{136}Xe/^{130}Xe$ gave a strong linear correlation (r = .98), suggesting that both components resided in the same mineral. Manuel *et al.* therefore proposed that both components were made in some earlier locale (such as a

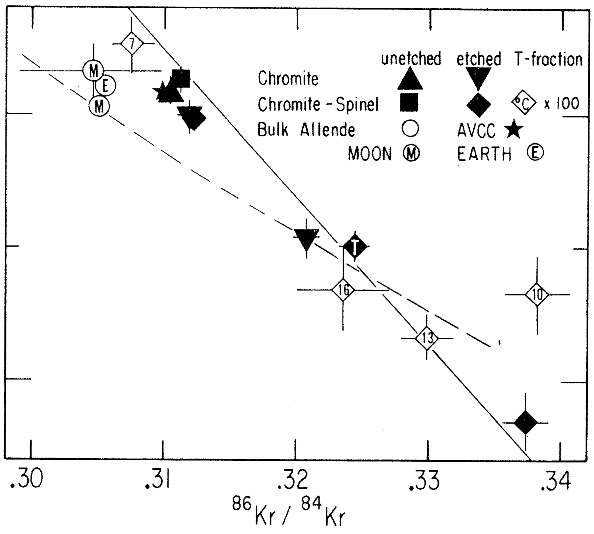

Fig. 6. Etched chromite samples are enriched in heavy Kr isotopes, due to either fission or mass fractionation. Apparent fission yields can be found from the *x*-intercept of the correlation line, on the assumption that the fission yield of shielded isotope ^{82}Kr is zero. The trapped Kr component in these samples presumably lies on a northwest extension of the correlation line; it cannot be derived from solar or terrestrial Kr by mass fractionation (dashed line).

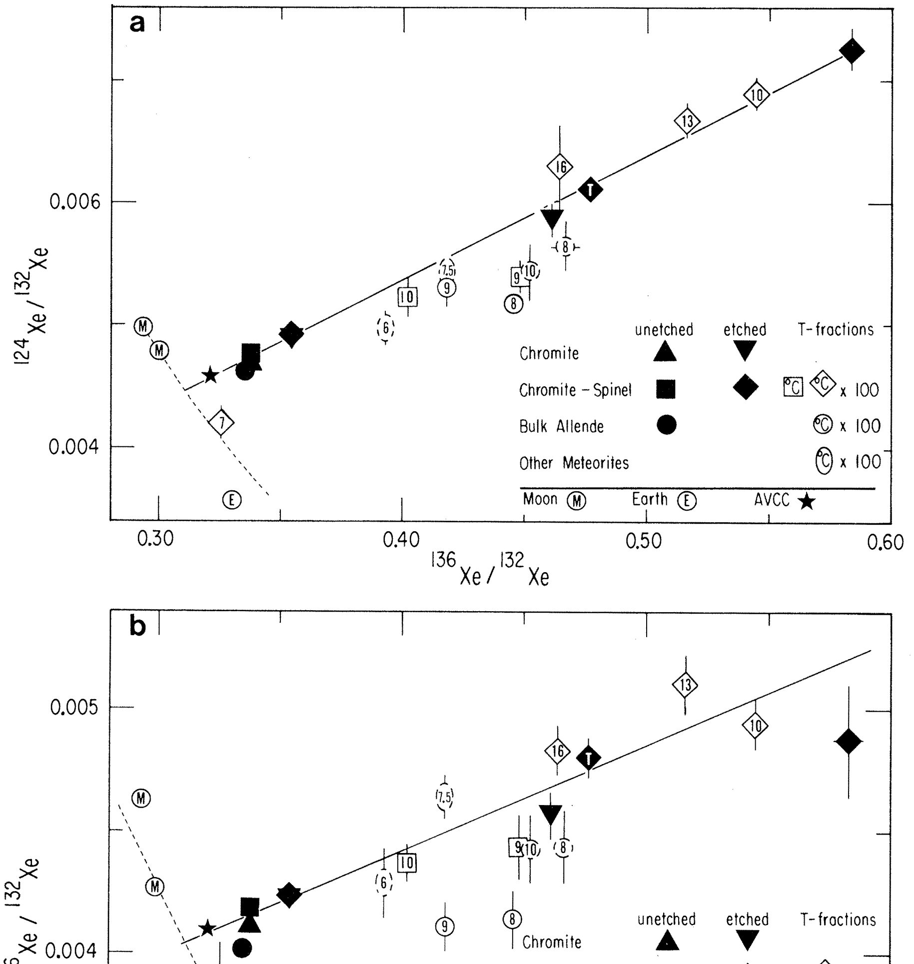

Fig. 7. (a and b) Xenon in etched Allende separates (chromite-carbon) shows pronounced enrichment in both light and heavy isotopes, which is masked in unetched samples by the normal trapped Xe in Q. As Q is etched away, samples move along the mixing line to the upper right. Light and heavy excesses do not seem to be strictly correlated in the other two host phases of fission Xe (Q, acid-soluble minerals) because temperature fractions from bulk meteorites and unetched Allende separates [open circles, ellipses, squares (*3, 13, 48*)] often plot below the mixing line. Apparently light and heavy excesses are independent components, not a single, homogenous component.

Table 7. Release of trapped and fission Xe on etching. The composition of the dissolved fraction was calculated from the difference between each residue and the preceding sample. For comparison, $(^{136}Xe/^{132}Xe)_{trapped} = 0.310$. Errors include only the uncertainties in isotopic ratios and weight fraction dissolved, not in ^{132}Xe concentration.

Treatment	^{132}Xe retained (%)		$^{136}Xe/^{132}Xe$			
			Residue		Dissolved	
	C	CS	C	CS	C	CS
None	100	100	0.338 ± .002	0.338 ± .002		
Aqua regia	43	47	0.354 ± .002	0.354 ± .003	0.326 ± .004	0.324 ± .005
HNO_3	13	12	0.461 ± .003	0.476 ± .002	0.310 ± .003	0.311 ± .004
Fuming HNO_3		6		0.583 ± .005		0.364 ± .005

supernova or the primitive sun) and then mixed in the gas phase before trapping. This point of view has also been adopted by Black (*14*).

Our data certainly support Manuel's observation of a close correlation between excess light and heavy Xe isotopes (Fig. 7). The Allende separates (filled symbols) show an excellent linear correlation, over an even wider range than Manuel's temperature fractions of bulk meteorites (dashed circles or ellipses). With the exception of ^{129}Xe, which is affected by the decay of extinct ^{129}I, all isotopes of Xe show such a correlation. This argues convincingly for the presence of only two Xe components in our residue: a normal trapped component and a component enriched in both the light and heavy isotopes.

The immediate, but too hasty, conclusion is that the light and heavy Xe excesses were homogenized before trapping in the meteoritic minerals. The correct conclusion is that the correlation is merely a mixing line between the gas contained in Q and the gas contained in chromite-carbon. This explains the correlation but leaves the question of previous homogenization unanswered. Note that because most of the Xe in Allende is contained in Q and chromite-carbon, this conclusion is also valid for the correlation observed in temperature fractions of the bulk meteorite (*13*).

We are thus left with the problem of deciding the relative merits of the two models: previous homogenization or in situ fission (coupled with some ad hoc mechanism for the light isotopes, such as mass fractionation during trapping in chromite). Two tests are available, each of which appears to favor the in situ fission model.

First, according to the homogenization model all three components would be trapped from a gas phase. Whether the trapping took place in one or two stages, all components should occupy identical crystallographic sites. Thus the normal trapped component, which comprises most of the gas in Q and even chromite, should be inseparable in a given mineral from the light and heavy excesses. In Table 7 we list the measured 136/132 ratios for the Xe remaining after various acid treatments, corresponding to increasingly complete removal of the normal trapped Xe. This ratio increases as Q dissolves, and the Xe in chromite-carbon becomes more dominant. Also listed are the calculated ratios for the Xe released under these treatments. The 136/132 ratio in the first $\sim$55 percent of the gas lies noticeably above the trapped ratio of 0.310, but declines to this value in the second fraction, comprising 30 to 35 percent of the gas. Such preferential early release of excess ^{136}Xe relative to normal trapped Xe is not consistent with the homogenization hypothesis. It would, of course, be interesting to see whether the light Xe excesses are also independent, but because of the extreme dilution by normal trapped Xe, the data are not accurate enough to tell.

The second test is to look for deviations from a strict correlation of light and heavy Xe. Previous homogenization predicts no such deviations, whereas in situ fission admits two possible reasons for such deviations: variations in the ratio of progenitor to light trapped Xe, and a radiation-damaged track leading to the fission Xe atom. For this test we use unetched samples (bulk meteorites and unetched separates) that contain the two remaining host phases of fission Xe: Q and HF,HCl-soluble minerals. Both phases also contain very large amounts of trapped Xe, and we must therefore limit ourselves to temperature fractions in which the heavy Xe is enhanced. Eight fractions of $^{136}Xe/^{132}Xe > 0.39$ were available: six from bulk meteorites [open circles or ellipses in Fig. 7 (*3, 5, 48*)], and two from the unetched separate 1CS [open squares (*48*)].

These temperature fractions from unetched samples do not all fall on the mixing line between Q and chromite-carbon. Fifteen of the 16 points in Fig. 7 fall below the line, 11 by more than 1 standard deviation. Part of the tendency toward a rough correlation must, of course, be due to the chromite-carbon, which still accounts for a major fraction of the heavy Xe (43 percent, compared to 24 and 33 percent for Q and acid-soluble minerals). Thus it appears that the ratio of light to heavy Xe is not constant in these minerals, contrary to the predictions of the homogenization hypothesis.

Although a case has been made for the independence of the heavy and light components, no direct evidence on this point is yet available for chromite, the phase richest in the light component. Etching experiments on chromite have not yet been performed, and a four-step heating experiment on 3CS2 failed to show a separation of light and heavy excesses (open diamonds in Fig. 7). No conclusions can be drawn from this negative result, because no separation would be expected unless chromite and carbon registered fission tracks, and retained them up to the temperatures where gas release becomes substantial. No data exist for chromite, but graphitic carbon is too conductive to record tracks.

Escape of Fission Fragments?

One major problem for the in situ hypothesis is the fine grain size of the gas-bearing minerals. The chromite-carbon grains in 3C1 are only about 0.3 μm in diameter, and although we have never recognized Q under the optical or scanning electron microscope, the absence of large particles suggests that it, too, is fine-grained (at least after our separation). But the range of fission fragments is 7 μm, and so if the tiny grains of Q and chromite-carbon were evenly dispersed throughout the meteorite, they should contain only 0.5 percent, not 67 percent, of the total fission Xe in the meteorite. In terms of the in situ hypothesis this requires that at least part of Q was clustered in aggregates larger than 7 μm and was surrounded by a layer of chromite-carbon thick enough to catch most of the escaping fission fragments. The chromite in our residue contains 14 percent of the Cr in the meteorite, and so if all the chromite has a uniform content of fission Xe, then 14 percent of the Cr in the meteorite must be clustered. Of course, if only a fraction of the chromite contains any fission Xe, then a correspondingly smaller fraction of the Cr must be clustered.

A microprobe search showed that much of the Cr is in fact dispersed. No clusters were found in an area where between one and six would be expected (depending on the chromite-carbon ratio in the cluster). The meteorite contains a uniform background of 0.5 ± 0.1 percent Cr_2O_3, compared to 0.52 percent in the bulk analysis (*24*), and the few rare, Cr-rich particles associated with sulfides are generally too small (< 3 μm). However, Larimer and Holdsworth (*49*) have found a number of promising features: large (15 to 20 μm) spheres of fine-grained chromite surrounding cores of Ni-rich pentlandite. They occur in large (70 to 80 μm) Mg- and Cr-rich olivine crystals, and are always connected to the matrix by fractures. Evidently chromite clusters of the right size do exist, although it has not yet been shown that they contain fission Xe, and are sufficiently abundant.

The Ne data further constrain the possible distribution of chromite. Chromite sample 3Cl, although depleted in the principal target elements Mg, Al, and Si, had a spallogenic ^{21}Ne concentration nearly equal to that of the bulk meteorite. This ^{21}Ne must have recoiled into the chromite-carbon from closely associated matrix or olivine no more than ~ 1 μm away [for a discussion of an analogous case see (*19*)]. The requirement that ~ 67 percent of the fission Xe be caught by chromite-carbon + Q makes this close association nearly impossible for matrix but tenable for olivine.

Fission Spectrum of Progenitor

Potentially, the new data can provide a much improved estimate of the fission spectrum of the progenitor, because the etched chromite separates contain a greater proportion of fission Xe than even the most enriched temperature fractions of bulk samples (dashed circle in Fig. 8). This advantage is offset by the even greater enhancement of the anomalous light Xe. Although obviously of different nuclear origin, it has remained closely coherent with the fission Xe in chromite (Fig. 7) and is therefore still too poorly defined for treatment as a separate component. Accordingly, we have treated the Xe in chromite as a mixture of fission Xe with a single trapped component. Since our analysis is based on only the 43 percent of the total Xe that resides in chromite, the neglected light component looms larger in our spectrum (Fig. 2 and Table 8) than in previous estimates (*1, 13, 50*).

The heavy isotopes show linear correlations on three-isotope plots such as Fig. 8, and can therefore be treated as simple binary mixtures of a trapped component similar to AVCC Xe (star) and a fission component off scale to the lower right. The fission yield of each isotope ^{i}Xe relative to ^{136}Xe is found from three-isotope plots of ^{130}Xe/^{i}Xe against ^{136}Xe/^{i}Xe (for example, Fig. 8). The intercept of the correlation line with the x-axis gives $(^{136}\mathrm{Xe}/^{i}\mathrm{Xe})_f$ of the pure fission component, on the assumption that the fission yield of the shielded isotope ^{130}Xe is zero and that all ^{130}Xe comes from a single trapped component. As noted above, we have misgivings about the latter assumption, because the light component accompanying the fission Xe is enriched in the isotopes 124 to 128 (Fig. 5) and probably contains some excess 130 as well. It should really be treated as a second trapped component. However, we have not yet found a satisfactory way to determine the isotopic composition of this component.

The trapped component in these samples must lie on the correlation line in Fig. 8, somewhere beyond the last point on the left. We assumed that it fell at the intercept of the correlation line with the mass fractionation curve of solar Xe. Its composition is given in Table 8.

Table 8. Fission Xe component in carbonaceous chondrites. The fission component was obtained from the intercept of three-isotope correlation lines, as discussed in the text. The composition of the trapped component was assumed to lie at the intersection of the correlation lines with a mass fractionation line passing through solar Xe [analysis BEOC 12001 on lunar soil (*45*)]. The isotopic ratios relative to ^{132}Xe were: ^{124}Xe (0.00446), ^{126}Xe (0.00404), ^{128}Xe (0.0814), ^{129}Xe (?), ^{130}Xe (0.162), ^{131}Xe (0.817), ^{132}Xe ($\equiv$ 1.00), ^{134}Xe (0.375), and ^{136}Xe (0.310).

Meteorite	$\frac{^{131}\mathrm{Xe}}{^{136}\mathrm{Xe}}$	$\frac{^{132}\mathrm{Xe}}{^{136}\mathrm{Xe}}$	$\frac{^{134}\mathrm{Xe}}{^{136}\mathrm{Xe}}$	Reference
Allende*	0.156 ±.011	0.092 ±.015	0.671 ±.017	This work
Renazzo	0.13 ±.10	0.16 ±.10	0.72 ±.04	(*1*)
Various	0.17 +.02 −.03	0.22 +.07 −.02	0.71 ±.01	(*50*)
Allende-800°C	0.21 ±.08	0.41 ±.10	0.71 ±.05	(*13*)

*An excess of the light isotopes was also present, with ratios ^{124}Xe/^{136}Xe = 0.010, ^{126}Xe/^{136}Xe = 0.004, and ^{128}Xe/^{136}Xe = 0.029.

The values for ^{131}Xe and especially ^{132}Xe of the fission component are lower than earlier estimates. A review of the older data shows that the temperature fractions of highest 136/132 ratio, which had the greatest influence on the slope, often had lower 130/132 ratios than our samples. Perhaps they contained an admixture of terrestrial-type Xe, from an atmospheric or meteoritic source (*1*). On the other hand, it is possible that our samples were enriched in ^{130}Xe from the light component. This would make our calculated fission yield of ^{132}Xe too low.

Origins: Supernova

One apparent advantage of a supernova is its ability to produce both light and heavy isotope anomalies. This ability is not crucial, though, as the light and heavy isotope anomalies are not strictly correlated. Another advantage of a supernova origin is the less severe constraint it places on the half-life. The progenitor need only survive long enough to condense on solid grains; if these grains are large enough (> 7 μm) and have a gentle enough temperature history, they will preserve the fission Xe indefintely, until their eventual incorporation in the meteorite (*17, 18*).

The second advantage may not be great, however. Cameron (*51*) has noted that an expanding supernova envelope is not a suitable milieu for condensation of grains. Temperatures are too high in the early stages of expansion, and they remain high, by shock heating, as the gas sweeps up interstellar material. Grains are likely to form only after the material has been

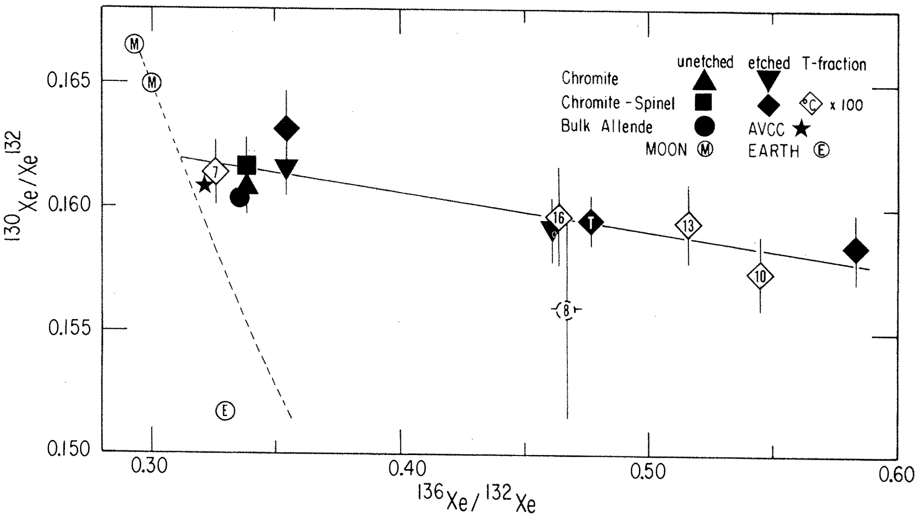

Fig. 8. Correlation plot for heavy Xe isotopes. Linear trend suggests that all Xe fractions are binary mixtures of a fission component (off scale at right) and a trapped Xe component lying near the intersection of the correlation line with the mass fractionation curve of solar Xe (dashed line). Fission yield of ^{132}Xe relative to ^{136}Xe is determined from x-intercept, on the assumption that the fission yield of ^{130}Xe is zero. Dashed circle (800°C) indicates the most fissiogenic composition observed by previous authors in stepwise heating experiments on bulk meteorites (*5*).

greatly diluted by ordinary interstellar matter. Judging from the conditions in old supernova remnants (Vela and the Cygnus Loop) this may take on the order of 10^5 years, and so the half-life must be at least as long. For comparison, if the progenitor is to reach the solar system in live form and in sufficient abundance, its half-life must be no less than 1.6×10^7 years (*52*).

Offsetting these advantages are several major difficulties. Chromite is a fairly rare mineral (0.6 percent of chondritic matter), located in the middle of the condensation sequence (*23, 53*). Unlike materials from the extreme ends of the condensation sequence, it cannot be separated from its complement of silicates, metal, and so forth by partial volatilization or condensation—in fact, the retention of anomalous Xe requires that the incorporation in the solar system was gentle. Thus it should be accompanied by a large complement of exotic metal, silicate, and so forth (about 100-fold for solar composition). Coming from an environment that produced large isotopic anomalies in Xe, these phases should display some isotopic anomalies of their own, particularly among *r*-process, *p*-process, and radiogenic isotopes. Yet careful studies of the Rb-Sr, U-Pb, ^{129}I-^{129}Xe, and ^{244}Pu-fission-Xe systems (*54*) have failed to show any anomalies attributable to an alien component. The only indications of such a component have been found in high-temperature, Ca,Al-rich inclusions from Allende, which show small variations in the isotopic compositions of O and Mg (*15, 16*). However, chromite and Q are associated with the matrix, not with these inclusions. A similar argument can be made for carbon, which, moreover, tends to associate with catalytic substrates (*55*).

A whole complex of problems is posed by the trapped noble gases, because the present study has shown that they reside in the same rare minerals as the fission Xe. Thus, if the fission Xe is extrasolar, the trapped gases are also. This leads to several problems.

First, the gases are too abundant for trapping in a tenuous supernova envelope or interstellar cloud. As we shall see, the high concentrations are an embarrassment even for an environment as dense as the solar nebula (pressure = 10^{-5} atm), making it necessary to assume that noble gases are five to eight orders of magnitude more soluble in Q than in the one mineral for which data exist, magnetite (*43*). The problem is aggravated by another five to ten orders of magnitude if the trapping is to take place in an interstellar cloud of correspondingly lower density. (Ion implantation obviates this difficulty, but produces the wrong elemental pattern unless the plasma fortuitously happens to have "planetary" composition; see Fig. 3.)

Second, it would be necessary to explain how a minor, exotic mineral came to be the dominant source of noble gases in this meteorite (and, by extension, other meteorites and planets). The elemental ingredients of this mineral were available in ample supply in the solar nebula, and so were noble gases.

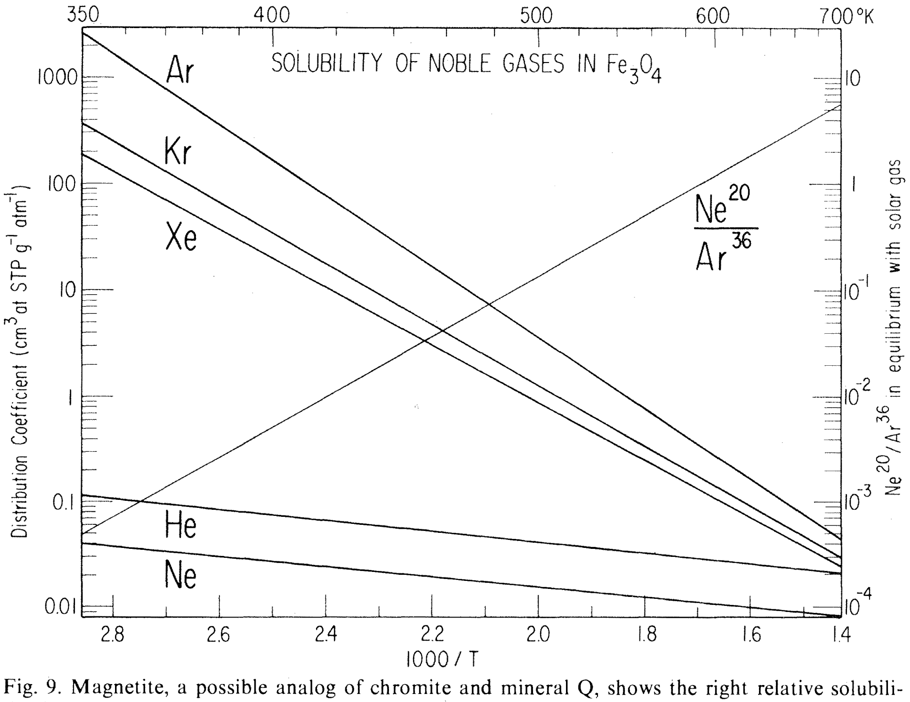

Fig. 9. Magnetite, a possible analog of chromite and mineral Q, shows the right relative solubilities for noble gases, although the absolute solubilities are too small by five to eight orders of magnitude (*43*). The light gases show a much smaller dependence of solubility on temperature, with the result that their ratios to heavy gases (for example, ^{20}Ne/^{36}Ar) are a sensitive function of temperature. Ratios of 4.3 and 0.05, as for chromite and Q, could be attained at 680°K and 460°K. It remains to be seen, however, whether chromite and Q indeed have the required high solvent capacity for noble gases.

Third, the isotopic composition of Ar, Kr, and Xe in Q is exceedingly close to that of the same elements in indisputably local samples: solar wind, the earth's atmosphere, and hundreds of meteorites. According to current views of stellar nucleosynthesis, these isotopic compositions are not universal, but vary appreciably from place to place. It would be surprising if the same exotic environment called upon to produce the fission Xe in Q were also able to supply large amounts of commonplace Ar, Kr, and Xe of essentially solar isotopic composition.

Further evidence on this point has been obtained by Tatsumoto (*56*). He showed that the Pb in Q has a very nonradiogenic isotopic composition, close to that of primordial solar-system Pb. Inasmuch as solar-system Pb represents only one of many possible compositions, this resemblance favors a local rather than an exotic origin of Q and its associated Xe.

Origins: Solar Nebula

Our main clue to the origin of fission Xe is the context in which it is found. From the present study, we have learned that it is contained in chromite-carbon and in mineral Q (presumably an iron-chromium sulfide). The higher concentration in Q suggests that Q, not chromite, was the original host phase of the progenitor. The greater lability of the fission Xe on etching suggests that the decay occurred in situ.

Both Q and chromite contain a characteristic suite of noble gases and trace elements, whose presence must be accounted for. We shall try to see whether conventional models of the solar nebula can explain the noble gas data, and at the expense of what assumptions.

A possible scenario for the formation of chromite and Q has been discussed in the companion article (*23*). Both minerals may have originated from Cr-bearing FeNi grains (*53*) that had condensed from the nebula between 1400° and 1200°K. On cooling, the grains reacted first with H_2O between 800° and 500°K to form chromite, and then with H_2S at 500° to 450°K to form Q. Carbon formed concurrently, perhaps by catalytic decomposition of CO on grain surfaces. It has been proposed (*42, 43*) that minerals forming in the nebula at such low temperatures would trap noble gases in dislocations or anion vacancies. We shall try to determine whether this process can account for the observed elemental and isotopic patterns in Q and chromite.

Elemental patterns. Noble-gas solubilities in chromite have not been measured. However, data are available for another spinel, Fe_3O_4 (*43*), which should be at least qualitatively similar to those for chromite, in view of the structural similarity of the two minerals and the high Fe^{3+} content of the chromite (*32*).

In magnetite, He and Ne have much smaller heats of solution than do Ar, Kr, and Xe: −2.4 and −2.2 kcal/mole, compared to −15.2, −13.0, and −12.5 kcal/mole (*43*). Consequently the solubilities of He and Ne rise by only a factor of 5 between 700° and 350°K, while the solubilities of the heavy gases rise by a factor of 10^5 (Fig. 9). The ratio of a light to a heavy gas in the solid is thus a sensitive function of temperature. This is illustrated in Fig. 9, which shows ^{20}Ne/^{36}Ar in magnetite equilibrated with a gas of solar ^{20}Ne/^{36}Ar ratio (*39*).

Qualitatively, this graph accounts for the Allende data. A high ^{20}Ne/^{36}Ar ratio (4.3) and low ^{36}Ar content (126×10^{-8} cm^3 STP/g), as for chromite 3CS4, would be expected for magnetite equilibrated at 680°K. A low ^{20}Ne/^{36}Ar ratio (0.05) and high ^{36}Ar content ($35{,}000 \times 10^{-8}$ cm^3 STP/g), as for Q (Table 6), would be expected for a temperature of 460°K.

Of course, these numbers are merely illustrative, because they refer to magnetite, not chromite, carbon, or Q. The temperature coefficients of solubility should be similar for all four minerals, because heats of solution, being dependent mainly on atomic properties of the noble gases, will always tend to show the same dichotomy between light and heavy noble gases. Absolute solubilities, on the other hand, depend on the abundance of anion vacancies and other lattice defects large enough to trap noble gases; these will differ from mineral to mineral. Indeed, we must assume that they are four to six and five to eight orders of magnitude higher in chromite-carbon and Q, respectively, than in magnetite, because the measured distribution coefficients for magnetite fail by this margin to account for the gas contents of the Allende samples, at a nebular pressure of 10^{-5} atm. (Alternatively, the pressure may have been four to eight orders of magnitude higher, but we consider this unlikely.)

It does not seem unreasonable that the abundance of trapping sites and solubilities should be higher by such factors. The saturation concentration for Xe in magnetite at 500°K is 3.1×10^{14} atom/g, or about 10^{-4} the number of O^{2-} vacancies and 3×10^{-8} the total number of O^{2-} sites (*43*). Sulfides have a double advantage over magnetite: normal (single) anion vacancies are large enough to accommodate Xe, and the tendency toward nonstoichiometric composition may yield a much higher abundance of vacancies, up to 0.1 or even greater. Chromite likewise has a tendency toward nonstoichiometry; moreover, the Allende chromite was highly disordered, as shown by the fact that lattice spacings contracted by about 1 percent after acid treatment.

Isotopic patterns. The principal trend to be explained is the strong fractionation in chromite, with its curious reversal at Xe. But the lack of such a fractionation in Q also needs to be explained.

The chromite trend essentially parallels the planetary noble gas pattern, which has been discovered, piece by piece, over the last 15 years (*6, 19, 36, 44, 45, 57*) and has remained unexplained during all this time. The obvious explanation, by Ockham's razor, is that the fractionation occurred during trapping of the gases. If the trapping indeed took place by crystal growth at low temperatures, then this process must be capable of fractionating isotopes, to a degree that differs from mineral to mineral.

No systematic experimental study of this process has been made. Magnetite apparently causes no isotopic fractionations greater than 1.5 percent per mass unit for Ne and Ar (*43*) and probably also Kr and Xe (*58*). Uranium oxides, on the other hand, fractionated Xe by up to 5 percent per mass unit at about 600°C (*59*). Both enrichments and depletions were observed.

Clearly, the problem cannot be regarded as solved until appropriate model experiments have been carried out on chromite, carbon, and Q. Such experiments will have important implications not only for meteorites, but also for the inner planets. Various authors have argued that the inner planets acquired their volatiles from a "thin veneer" of carbonaceous chondrite material (*60*). Now that three host minerals of noble gases have been identified, it should be possible to arrive at a better understanding of this process.

Summary

We have isolated the host phase of an enigmatic Xe component in the Allende meteorite, which may have been produced by spontaneous fission of an extinct superheavy element. The fission Xe resides in a minor fraction comprising 0.5 percent of the meteorite, and consisting of chromite, an unknown Cr,Fe-mineral (Q, possibly a sulfide), and amorphous carbon. More than one-half of the trapped noble gases in the meteorite are located in these three minerals. Mineral Q contains gases of essentially solar isotopic composition (most of the Ar, Kr, and Xe, and some He and Ne) while chromite-carbon contains gases of strongly fractionated isotopic composition (Ne, Ar, Kr enriched in heavy isotopes, and Xe enriched in light isotopes). Both gas components, and the progenitor of the fission Xe, may have been trapped in these minerals when they formed from the solar nebula.

References and Notes

1. J. H. Reynolds and G. Turner, *J. Geophys. Res.* **69**, 3263 (1964).
2. R. O. Pepin, in *Origin and Evolution of Atmospheres and Oceans*, P. J. Brancazio and A. G. W. Cameron, Eds. (Wiley, New York, 1964), p. 191; in *Origin and Distribution of the Elements*, L. H. Ahrens, Ed. (Pergamon, Oxford, 1968), p. 379.
3. H. Funk, F. Podosek, M. W. Rowe, *Geochim. Cosmochim. Acta* **31**, 1721 (1967); M. W. Rowe, *ibid.* **32**, 1317 (1968).
4. P. K. Kuroda and O. K. Manuel, *Nature (London)* **227**, 1113 (19→ O. K. Manuel, R. J. Wright, D. K. Miller, P. K. Kuroda, *J. Geophys. Res.* **75**, 5693 (1970); P. K. Kuroda, J. N. Beck, D. W. Efurd, D. K. Miller, *ibid.* **79**, 3981 (1974); P. K. Kuroda, R. D. Sherrill, D. W. Efurd, J. N. Beck, *ibid.* **80**, 1558 (1975).
5. O. K. Manuel, R. J. Wright, D. K. Miller, P. K. Kuroda, *Geochim. Cosmochim. Acta* **36**, 961 (1972).
6. K. Marti, *Earth Planet. Sci. Lett.* **3**, 243 (1967); O. Eugster, P. Eberhardt, J. Geiss, *ibid.*, p. 249; F. A. Podosek, J. C. Huneke, D. S. Burnett, G. J. Wasserburg, *ibid.* **10**, 199 (1971).
7. E. C. Alexander, Jr., R. S. Lewis, J. H. Reynolds, M. C. Michel, *Science* **172**, 837 (1971); K. F. Flynn, B. Srinivasan, O. K. Manuel, L. E. Glendenin, *Phys. Rev. C* **6**, 2211 (1972); G. W. Wetherill, *Phys. Rev.* **92**, 907 (1953).

→ B. Srinivasan, E. C. Alexander, Jr., O. K. Manuel, D. E. Troutner, *Phys. Rev.* **179**, 1166 (1969).

9. E. Anders and D. Heymann, *Science* **164**, 821 (1969).

→ M. Dakowski, *Earth Planet. Sci. Lett.* **6**, 152 (1969).

11. G. T. Seaborg, *Annu. Rev. Nucl. Sci.* **18**, 53 (1968).
12. E. Anders and J. W. Larimer, *Science* **175**, 981 (1972).
13. O. K. Manuel, E. W. Hennecke, D. D. Sabu, *Nature (London) Phys. Sci.* **240**, 99 (1972).

→ D. C. Black, *Nature (London)* **253**, 417 (1975).

15. R. N. Clayton, L. Grossman, T. K. Mayeda, *Science* **182**, 485 (1973).
16. T. Lee and D. A. Papanastassiou, *Geophys. Res. Lett.* **1**, 225 (1974); P. Eberhardt, *Earth Planet. Sci. Lett.* **24**, 182 (1974).

→ D. D. Clayton, *Astrophys. J.* **199**, 765 (1975).

18. W. M. Howard, M. Arnould, J. W. Truran, *Astrophys. Space Sci.* **36**, L1 (1975).
19. P. M. Jeffery and E. Anders, *Geochim. Cosmochim. Acta* **34**, 1175 (1970).
20. G. F. Herzog and E. Anders, *Earth Planet. Sci. Lett.* **24**, 173 (1974).
21. E. A. Carver, thesis, University of Chicago (1974).
22. See R. S. Lewis and E. Anders [*Proc. Natl. Acad. Sci. U.S.A.* **72**, 268 (1974)] for extraction and purification of noble gases and B. Srinivasan [*Geochim. Cosmochim. Acta Suppl. 4* (1973), p. 2049] for separation of different noble gases, mass spectrometric analyses, data reduction, and errors.
23. E. Anders, H. Higuchi, J. Gros, H. Takahashi, J. W. Morgan, *Science* **190**, 1262 (1975).
24. R. S. Clarke, Jr., E. Jarosewich, B. Mason, J. Nelen, M. Gómez, J. R. Hyde, *Smithson. Contrib. Earth Sci. No. 5* (1970).
25. A sample of Allende was ground in a ball mill for 16 hours, suspended in 20 parts of water, and stirred on a magnetic stirrer. The magnetic fraction clinging to the stirring bar was resuspended in water and purified by repeating the above cycle. The final sample 1M (2.5 percent of the total) was only modestly enriched in metal, judging from an instrumental neutron activation analysis: Ni, 7.63 percent; Fe, 23 percent; and Cr, 0.44 percent (*30*). A sulfide concentrate was prepared by flotation with potassium ethyl xanthate and *n*-amyl alcohol at *p*H 7 to 9. Although the float fraction was reprocessed twice, the enrichment was rather poor, because even silicate particles containing only a small speck of sulfide floated quite persistently. The final sample 1SU was still contaminated with ~ 70 percent transparent material, and contained 3.25 percent Ni, 27 percent Fe, and 0.63 percent Cr.
26. No blank corrections were applied because only negligible amounts of gas, of atmospheric composition, were obtained in the blank runs. For the samples in Table 1, the extraction time was 15

minutes at 1600°C, yielding a ^{132}Xe blank of 0.48×10^{-12} cm^3 STP. For the samples in Tables 4 and 5, the extraction time was 30 minutes. Blanks at 1600°C (or 25°C) gave the following results, in 10^{-12} cm^3 STP: ^{84}Kr, 3.0 (0.3) and ^{132}Xe, 0.44 (0.10). The background of the mass spectrometer was negligible in the Kr-Xe region, except at masses 78 and 80. Corrections were applied at these masses, and one-half of the correction was included in the error. All isotopic ratios were corrected for mass discrimination, and the uncertainty in this correction was likewise included in the error. The errors in this article are 1 standard deviation.
27. E. Mazor and E. Anders, *Geochim. Cosmochim. Acta* **31**, 1441 (1967); H. W. Weber, H. Hintenberger, F. Begemann, *Earth Planet. Sci. Lett.* **13**, 205 (1971).
28. X-ray diffraction work indicated the presence of spinel and of chromite. The spinel x-ray diffraction pattern matched perfectly with the pattern obtained by R. N. Clayton (private communication) on spinels separated by heavy liquids from Allende's white inclusions. The chromite x-ray diffraction pattern was not as sharp as the spinel pattern, which indicated smaller particle size. Curiously, treatment of the chromite with HNO_3 or aqua regia at ~ 80°C caused a decrease in the spacings by ~1 percent. An extensive but unsuccessful search was made for any additional lines and also for lines removed by the HNO_3 and aqua regia treatments.
29. L. Grossman and J. W. Larimer, *Rev. Geophys. Space Phys.* **12**, 71 (1974).
30. J. Gros, unpublished work.
→ I. A. Breger, P. Zubovic, J. C. Chandler, R. S. Clarke, *Nature (London)* **236**, 155 (1972).
32. D. Virgo, private communication.
33. L. Gmelin, *Handbuch der Anorganischen Chemie*, vol. 52, *Chrom* (Verlag Chemie, Weinheim, ed. 8, 1962), part B.
34. E. R. DuFresne and E. Anders, *Geochim. Cosmochim. Acta* **26**, 251 (1962).
35. E. Vilcsek and H. Wänke, *Z. Naturforsch.* **20a**, 1282 (1965).
→ E. Mazor, D. Heymann, E. Anders, *Geochim. Cosmochim. Acta* **34**, 781 (1970).
37. This archaic term has been restored to active use by D. A. Papanastassiou and G. J. Wasserburg [*Earth Planet. Sci. Lett.* **11**, 37 (1971)]. They use it to designate a minor glassy phase in lunar rocks that is highly enriched in trace elements such as Rb and U.
38. R. S. Lewis, J. Gros, E. Anders, unpublished work.
→ A. G. W. Cameron, *Space Sci. Rev.* **15**, 121 (1973).
40. D. Heymann and A. Yaniv, *Geochim. Cosmochim. Acta Suppl. 1* (1970), p. 1247.
41. K. Marti, *Science* **166**, 1263 (1969).
→ J. W. Larimer and E. Anders, *Geochim. Cosmochim. Acta* **31**, 1239 (1967).
→ M. S. Lancet and E. Anders, *ibid.* **37**, 1371 (1973).
44. D. C. Black and R. O. Pepin, *Earth Planet. Sci. Lett.* **6**, 395 (1969); D. C. Black, *Meteoritics* **4**, 260 (1969); *Geochim. Cosmochim. Acta* **36**, 377 (1972).
45. P. Eberhardt, J. Geiss, H. Graf, N. Grögler, M. D. Mendia, M. Mörgeli, H. Schwaller, A. Stettler, *Geochim. Cosmochim. Acta Suppl. 3* (1972), p. 1821.
46. O. Eugster, P. Eberhardt, J. Geiss, *Earth Planet. Sci. Lett.* **3**, 249 (1967); F. A. Podosek, J. C. Huneke, D. S. Burnett, G. J. Wasserburg, *ibid.* **10**, 199 (1971).
47. R. S. Lewis, *Geochim. Cosmochim. Acta* **39**, 417 (1975).
48. B. Srinivasan and R. S. Lewis, unpublished work.
49. J. W. Larimer and E. F. Holdsworth, private communication.
50. D. L. Phinney, thesis, University of Minnesota (1971).
51. A. G. W. Cameron, in *Proceedings of a Symp. on the Dusty Universe*, Cambridge, Mass., 1973.
52. D. N. Schramm, *Nature (London)* **233**, 258 (1971).
53. L. Grossman and E. Olsen, *Geochim. Cosmochim. Acta* **38**, 1973 (1974).
54. C. M. Gray, D. A. Papanastassiou, G. J. Wasserburg, *Icarus* **20**, 213 (1973); F. A. Podosek and R. S. Lewis, *Earth Planet. Sci. Lett.* **15**, 101 (1972); M. Tatsumoto, D. M. Unruh, G. DesBorough, *Geochim. Cosmochim. Acta*, in press.
55. E. Anders, R. Hayatsu, M. H. Studier, *Science* **182**, 781 (1973).
56. M. Tatsumoto, private communication.
57. P. Signer and H. E. Suess, in *Earth Science and Meteoritics*, J. Geiss and E. D. Goldberg, Eds. (North-Holland, Amsterdam, 1963), p. 241.
58. M. S. Lancet, thesis, University of Chicago (1972).
59. W. B. Lewis, *At. Energy Can. Ltd. AECL Rep. 1402* (1961).
60. H. C. Urey, in *L. Farkas Memorial Volume* (Research Council of Israel, Jerusalem, 1952), p. 3; N. Kokubu, T. Mayeda, H. C. Urey, *Geochim. Cosmochim. Acta* **21**, 247 (19→ E. Anders, *Acc. Chem. Res.* **1**, 289 (1968); K. K. Turekian and S. P. Clark, Jr., *Earth Planet. Sci. Lett.* **6**, 346 (1969).
61. We are most grateful to I. Steele for his unstinting assistance in the use of the electron microprobe. We also thank R. N. Clayton, L. Grossman, T. K. Mayeda, and J. V. Smith for fruitful discussions and assistance in the x-ray diffraction work. Special thanks go to J. Ito, whose work first indicated the chemical nature of our residue. Our rare gas data reduction computer program was provided by J. H. Reynolds' rare gas mass spectrometry group in Berkeley. We thank K. M. O'Connell for typing the manuscript under extreme pressure, and Rudolph Banovich for preparation of the drawings. Klaus Keil kindly provided the meteorite sample. This work was supported in part by NASA grant NGL 14-001-010.

23 July 1975; revised 31 October 1975

Extinct Superheavy Element in the Allende Meteorite

Element 115 (or 114, 113) may have been present in a rare chromium mineral when the meteorite formed.

Edward Anders, H. Higuchi, Jacques Gros,
H. Takahashi, John W. Morgan

Eleven years after Reynolds and Turner (*1*) discovered excess $^{131-136}$Xe in the Renazzo meteorite, the source of the xenon remains unidentified. Presumably it is an unknown transuranium element that decayed by spontaneous fission. But none of the known transuranium elements, including extinct 82×10^6 year ^{244}Pu, produces xenon of this peculiar isotopic composition (*2–4*).

Only one clue to the chemical nature of this element has been available: its volatility. The fission-produced xenon occurs solely in primitive, volatile-rich meteorites, and Anders and Heymann (*5*) therefore suggested that the progenitor of the xenon was to be sought among elements 112 to 119, the only transuranium elements expected to be volatile (*6*). Anders and Larimer (*7*) later revised the range to 111 to 116, after estimating the condensation temperatures of superheavy elements in the solar nebula.

These arguments were based on the distribution of the free element between nebular gas and meteoritic solids, as inferred from the abundance of fission xenon in meteorites of different accretion temperature. The obvious next step (*5, 7*) was to study the distribution of fission xenon among various meteoritic minerals, and compare it with that of other trace elements. This distribution might reveal some further chemical properties of the progenitor, such as oxidation state, ionic radius, and geochemical character (that is, tendency to form oxides, sulfides, or free metal).

This experiment finally became feasible with the isolation of a mineral fraction from the Allende C3 chondrite that was enriched nearly 200-fold in fission xenon, as described in the companion article by Lewis *et al.* (*8*). A mass spectrometric study of the noble gases in this fraction is reported in (*8*). This article describes a radiochemical study of the same fraction.

Results

We have analyzed seven Allende samples for 26 trace elements by radiochemical neutron activation analysis, using

---

Edward Anders is Horace B. Horton Professor at the Enrico Fermi Institute and Department of Chemistry, University of Chicago, Chicago, Illinois 60637. H. Higuchi, formerly a research associate at the University of Chicago, is now at the Nippon Bunseki Center, Itabashi, Tokyo, Japan. Jacques Gros and H. Takahashi are research associates and John W. Morgan is senior research associate at the Enrico Fermi Institute and Department of Chemistry, University of Chicago.