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# Terrestrial-type xenon in sulfides of the Allende meteorite

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Two FeS-rich samples from the Allende (C3V) chondrite were analyzed for Xe and Kr. One sample was irradiated in a neutron flux to generate a tracer isotope of Xe by the $^{130}Te(n,\gamma 2\beta^-)^{131}*Xe$ reaction. The experiment was designed to use this tracer isotope to identify the temperature at which gases were released from within the sulfide. When the sulfide melted at 950°C, the Xe released was terrestrial in isotopic composition, except for enrichments from spallogenic and radiogenic components. It is concluded that terrestrial-type Xe, Xe-T, was a primordial component that was dominant in the inner Fe, S-rich region of the solar nebula.

## INTRODUCTION

Xenon isotopes in meteorites have provided a wealth of information on the origin and early history of the Solar System, including the first evidence for decay products from two extinct radioactive nuclides, 16 my $^{129}I$ (Reynolds, 1960a) and 82 my $^{244}Pu$ (Rowe and Kuroda, 1965), and the first evidence of an isotopically distinct, primordial element in carbonaceous chondrites, AVCC Xe (Reynolds, 1960a, 1960b; Eugster *et al.*, 1967).

Another component, initially identified as *in situ* fission products (Reynolds and Turner, 1964), became known as CCF Xe, Carbonaceous Chondrite Fission Xenon (Pepin, 1968; Reynolds, 1970). Later it was found that excess $^{124}Xe$ and $^{126}Xe$ always accompany the excess $^{134}Xe$ and $^{136}Xe$ of CCF Xe (Manuel *et al.*, 1972), and it was noted that these may be from a supernova explosion, where $^{124}Xe$ and $^{126}Xe$ are produced by the p-process; $^{134}Xe$ and $^{136}Xe$ by the r-process (Burbidge *et al.*, 1957). Xe enriched in r- and p-products of nucleosynthesis was labeled Xe-X (Manuel *et al.*, 1972).

Lewis *et al.* (1975) enriched the host phase of Xe-X in acid-residues of Allende and attributed the excess $^{134,136}Xe$ to superheavy-element fission (Anders *et al.*, 1975). However, their data confirmed the presence of excess $^{124}Xe$ and $^{126}Xe$ and also revealed linear correlations between elemental abundances of He and Ne with isotopic ratios of Xe, Kr and Ar, as expected for a mixture of noble gases from different regions of a supernova (Manuel and Sabu, 1975, 1977): a) *Isotopically "strange" Ar, Kr and Xe (Xe-X) from the outer stellar layers where low-Z elements like He and Ne were abundant*; b) *Isotopically "normal" Xe, Kr and Ar from the stellar interior where fusion reactions depleted He and Ne.* Figure 6 of Oliver *et al.* (1981) shows how a supernova could produce such chemical and isotopic heterogeneities in the protoplanetary nebula.

Srinivasan and Anders (1978) later found Xe-S, enriched in $^{128-132}Xe$, a complimentary component to Xe-X. These isotopes are produced by slow neutron capture before a star reaches the supernova stage (Burbidge *et al.*, 1957). Nucleogenetic isotopic anomalies were also found in different elements of Allende and other meteorites (e.g., reviews by Begemann, 1980, 1993; Lavrukhina, 1980; Clayton, 1981; Lee, 1988). The nucleosynthesis hypothesis for Xe-X was finally adopted (Lewis *et al.*, 1983a, 1983b; Lugmair *et al.*, 1983; Swart, 1983), but not the suggestion of local element synthesis (Manuel and Sabu, 1975,

1977, 1981; Lavrukhina, 1980) and formation of the solar system from a single supernova.

Isotopic anomalies are instead attributed to remote nucleosynthesis and to interstellar grains that carried these alien products into the solar system. For Xe-X and Xe-S, these are reported to be interstellar diamonds with a mean diameter of ≈26 Å (Lewis *et al.*, 1987; Lewis and Anders, 1988) and silicon carbide with a mean diameter of 0.12 μm (Tang and Anders, 1988). However, isotopic anomalies occur on a much larger scale. Solar, terrestrial and AVCC Xe are isotopically distinct. These types of Xe are from the sun (Eberhardt *et al.*, 1972), in the atmospheres of Earth and Mars, ≈1–1.5 AU from the sun (Nier, 1950; Becker and Pepin, 1984), and in carbonaceous chondites, even further from the sun (Eugster *et al.*, 1967).

FeS mineral separates of Allende were selected for analysis to seek additional evidence for correlated chemical and isotopic heterogeneities because a) Allende is a member of the Type-III carbonaceous chondrites, which have the highest abundances of FeS (Kaplan and Hulston, 1966), b) FeS is composed of elements that are abundant in the inner planets, c) FeS contains readily measurable quantities of trapped Xe, and d) other mineral separates of Allende contain several isotopically anomalous elements also bearing the signature of supernova nucleosynthesis (Begemann, 1993).

In discussing our results, we use AVCC-Xe, Xe-S, Xe-X and Xe-T to represent Xe (Eugster *et al.*, 1967) in bulk carbonaceous chondrites, in SiC (Tang and Anders, 1988), in diamonds (Lewis and Anders, 1988), and in the terrestrial planets (Ozima and Podosek, 1983; Becker and Pepin, 1984).

## Samples and Procedures

### *Samples*

Sulfides used in this work came from University of Missouri-Rolla sample # M68. This single piece of the Allende meteorite (C3V), weighing 127.1 g, was purchased from the American Meteorite Laboratory in 1985. After mechanical removal of a black fusion crust, the remaining sample weighed 120.0 g. This was broken into smaller pieces with a hammer and placed in a SPEC Mill/Mixer, with sample vial and balls made of zirconia ceramic, to break into smaller fragments. The Mill/Mixer was operated for 2–3 seconds, fragments ≤1 mm were removed, additional sample was added, and then the process repeated until all the sample had been reduced to ≤1 mm. Fragments were then put in distilled water, ultrasonicated, and disaggregated by alternate exposure to liquid nitrogen, then to boiling water, and then back to ultrasonication. The suspension of fine particles was decanted from time to time. This treatment was repeated some 50 cycles until ultrasonication was no longer effective. The coarse fraction (≥50 μm) was further treated with heavy liquid, subdivided by sieving, and sulfide particles were handpicked from the high density fraction under a microscope. EDX analysis on early separates identified grains of troilite or pentlandite with the atomic ratio, Fe/S ≈ 1. Visual properties of these grains were used to identify additional sulfide grains under the microscope. This tedious process was halted when a total of 60.0 mg of sulfides had been recovered. Half of this sample, consisting of visually pure sulfides of ≈100–200 μm in size, was encapsulated in an evacuated quartz vial for neutron irradiation at the University of Missouri Research Reactor, MURR Graphite Reflector, Row 2, where (total flux/epithermal flux) was ≈12, the estimated temperature of the sample during irradiation was 36°–39°C, and the integrated neutron flux was $1.69 \times 10^{18}$ n/cm$^2$.

### *Experimental procedures*

These two sulfide mineral separates, each weighing 30.0 mg, were wrapped in Al foil and mounted in individual side-arm chambers of the gas extraction system. Before gas extraction, the samples were heated to ≈150°C for several days to expel adsorbed gases and to help reduce the pressure to about $10^{-9}$ torr. The unirradiated sample was analyzed first. This was dropped into a previously out-gassed stainless steel tube. Gases from the sample were collected and analyzed as it was heated stepwise to progressively higher temperatures: 600°C, 800°C, 950°C, 1000°C, and

1050°C. Each temperature was maintained for 40 minutes. Gases were cleaned twice by exposure to Ti-Zr getters at about 850°C. After the second scrubbing, the Ti-Zr was cooled to room temperature, and the Kr and Xe were trapped by adsorption on activated charcoal cooled with a slurry of solid $CO_2$ and ethanol. The lighter noble gases were pumped away, and then Kr was released from the charcoal by raising the temperature to –38.9°C, the freezing point of Hg. Kr was analyzed in a Reynolds-type, 11.4 cm sector noble gas mass spectrometer, operating in the static mode. Following analysis, Kr was pumped away and Xe was admitted with the charcoal at 200°C. Standard air pipettes of Kr and Xe gases were analyzed before and after the samples to obtain the sensitivity and mass discrimination of the instrument. Procedural blanks were $5.5 \times 10^{-13}$ cc STP for $^{132}$Xe and $4.3 \times 10^{-12}$ cc STP for $^{84}$Kr. More detailed information on gas extraction, purification and data acquisition is available elsewhere (Li and Manuel, 1994).

## Results and Discussion

The results of our analyses on the abundances of Kr and Xe and the isotopic compositions of Xe are shown in Table 1. Data tabulated there have been corrected for mass discrimination and procedural blanks. The errors in Table 1 represent one standard deviation ($\sigma$). The suffix (1) is used to designate the non-irradiated sulfide sample; the suffix (2) to indicate the irradiated one. At the bottom of the table are similar data for atmospheric (Ozima and Podosek, 1983), solar (Eberhardt *et al.*, 1972) and average carbonaceous chondrite (Eugster *et al.*, 1967) gases. Xenon isotopes are normalized to $^{132}$Xe, the most abundant Xe isotope in air.

The amounts of $^{84}$Kr and $^{132}$Xe released at each temperature are shown on the left side of Table 1. The amounts of $^{84}$Kr released were only slightly above that of the procedural blank for all extraction temperatures of FeS(1) and for the higher extraction temperatures of FeS(2). Only upper limits are shown when the gas released was less than 6 times that of the procedural blank. The next column shows values of the $^{84}$Kr/$^{132}$Xe ratio. This ratio is lower for gases from Allende sulfides than that in solar-wind implanted gases, much lower than that in air, and closest to that observed in carbonaceous chondrites. Several of the upper limit values of the $^{84}$Kr/$^{132}$Xe ratio in FeS(1) are even lower than that of AVCC.

*Release patterns for primordial, pile-produced and radiogenic xenon*

The unirradiated sample, FeS(1), released significantly less gas than the irradiated one, FeS(2). We suspect that the higher noble gas content of FeS(2) reflects the fact that only larger, more pure sulfide particles, ≈100–200 $\mu$m in size, were selected for irradiation. From a comparison of the $^{132}$Xe released from irradiated and unirradiated samples of meteoritic troilite (Niemeyer, 1979) and terrestrial tellurides (Kirsten *et al.*, 1967; Alexander *et al.*, 1969; Richardson *et al.*, 1986; Lin *et al.*, 1988; Lee *et al.*, 1991; Bernatowicz *et al.*, 1993), we find no systematic enrichment of noble gases in the irradiated samples. We will show later that FeS(1) contains more radiogenic $^{129}$Xe than FeS(2), as expected if the FeS(1) contains more high temperature silicates than FeS(2).

The release of $^{132}$Xe from the FeS(1) and FeS(2) is shown in Figs. 1a and 1b, respectively. For both samples, the largest amount of $^{132}$Xe was released at 950°C. The more pronounced peak release of $^{132}$Xe in the 950°C fraction of FeS(2) (see Fig. 1b) argues against surface adsorbed gases as an explanation for the higher amounts of noble gases in this sample. Adsorbed Xe would be selectively released at low temperature, but it can be seen in Fig. 1 that the fraction of Xe released below 950°C from FeS(2) (Fig. 1b) is even smaller than that from FeS(1) (Fig. 1a).

The amounts of pile-produced $^{131}$*Xe from $^{130}$Te(n,$\gamma 2\beta^-$) and $^{128}$*Xe from $^{127}$I(n,$\gamma\beta^-$) in each temperature fraction were calculated by subtracting values of the $^{131}$Xe/$^{132}$Xe and $^{128}$Xe/$^{132}$Xe ratios in FeS(1) from those in the irradiated sample, FeS(2), respectively. The release patterns of $^{131}$*Xe and $^{128}$*Xe from FeS(2) are shown in Fig. 2, in the

*Table 1. Abundances of Kr and Xe and isotopic composition of Xe in sulfide samples*

| Sample | $^{84}$Kr | $^{132}$Xe | $^{84}$Kr/$^{132}$Xe | Isotopic composition | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| (°C) | $\times 10^{-10}$ ccSTP/g | | | $^{124}$Xe | $^{126}$Xe | $^{128}$Xe | $^{129}$Xe | $^{130}$Xe | $^{131}$Xe | $^{132}$Xe | $^{134}$Xe | $^{136}$Xe |
| FeS(1) | | | | | | | | | | | | |
| 600 | ≤2.32 | 5.06 | ≤0.46 | 0.368 | 0.331 | 7.349 | 100.58 | 15.24 | 78.65 | 100.0 | 38.81 | 32.79 |
| | | | | ±0.026 | ±0.010 | ±0.111 | ±0.98 | ±0.13 | ±0.73 | | ±0.41 | ±0.32 |
| 800 | ≤0.82 | 7.67 | ≤0.11 | 0.337 | 0.347 | 7.306 | 99.03 | 14.99 | 78.43 | 100.0 | 38.98 | 32.91 |
| | | | | ±0.017 | ±0.023 | ±0.098 | ±0.97 | ±0.19 | ±0.81 | | ±0.35 | ±0.31 |
| 950 | ≤1.11 | 9.39 | ≤0.12 | 0.343 | 0.355 | 7.264 | 109.47 | 14.75 | 78.67 | 100.0 | 38.84 | 33.20 |
| | | | | ±0.014 | ±0.007 | ±0.070 | ±0.92 | ±0.18 | ±0.73 | | ±0.41 | ±0.32 |
| 1000 | ≤0.13 | 1.07 | ≤0.12 | 0.435 | 0.348 | 7.585 | 205.14 | 15.42 | 79.11 | 100.0 | 38.32 | 32.89 |
| | | | | ±0.028 | ±0.042 | ±0.170 | ±2.65 | ±0.26 | ±0.93 | | ±0.34 | ±0.25 |
| 1050 | ≤0.59 | 1.42 | ≤0.42 | 0.452 | 0.387 | 8.303 | 290.09 | 15.03 | 80.80 | 100.0 | 39.04 | 32.90 |
| | | | | ±0.011 | ±0.037 | ±0.168 | ±3.06 | ±0.21 | ±1.12 | | ±0.55 | ±0.46 |
| FeS(2) | | | | | | | | | | | | |
| 600 | 13.9 | 8.29 | 1.68 | 0.359 | 0.342 | 11.259 | 98.94 | 15.08 | 81.22 | 100.0 | 39.16 | 33.03 |
| | | | | ±0.018 | ±0.015 | ±0.234 | ±1.74 | ±0.33 | ±1.38 | | ±0.52 | ±0.61 |
| 800 | 31.6 | 16.22 | 1.95 | 0.317 | 0.329 | 8.504 | 98.67 | 15.18 | 80.29 | 100.0 | 38.60 | 32.83 |
| | | | | ±0.009 | ±0.012 | ±0.099 | ±0.84 | ±0.16 | ±0.71 | | ±0.34 | ±0.27 |
| 950 | 73.8 | 32.97 | 2.24 | 0.357 | 0.342 | 7.930 | 100.95 | 15.14 | 80.38 | 100.0 | 38.63 | 33.12 |
| | | | | ±0.007 | ±0.007 | ±0.070 | ±0.86 | ±0.17 | ±0.65 | | ±0.33 | ±0.26 |
| 1000 | ≤3.7 | 3.83 | ≤0.97 | 0.373 | 0.314 | 9.163 | 121.89 | 14.93 | 79.53 | 100.0 | 38.80 | 32.64 |
| | | | | ±0.019 | ±0.019 | ±0.128 | ±0.94 | ±0.11 | ±0.76 | | ±0.28 | ±0.33 |
| 1050 | ≤2.3 | 2.88 | ≤0.80 | 0.338 | 0.360 | 9.792 | 142.80 | 15.65 | 81.95 | 100.0 | 38.87 | 33.54 |
| | | | | ±0.026 | ±0.014 | ±0.194 | ±1.41 | ±0.20 | ±0.93 | | ±0.46 | ±0.41 |
| Air[a] | | | 28.8 | 0.354 | 0.330 | 7.14 | 98.32 | 15.14 | 78.91 | 100.0 | 38.79 | 32.94 |
| Solar[b] | | | 6.2 | 0.478 | 0.427 | 8.31 | 104.82 | 16.50 | 82.28 | 100.0 | 36.94 | 29.99 |
| AVCC[c] | | | 1.4 | 0.459 | 0.410 | 8.20 | | 16.08 | 81.70 | 100.0 | 38.20 | 32.10 |

[a]*Ozima and Podosek (1983).*
[b]*Eberhardt et al. (1972).*
[c]*Eugster et al. (1967).*

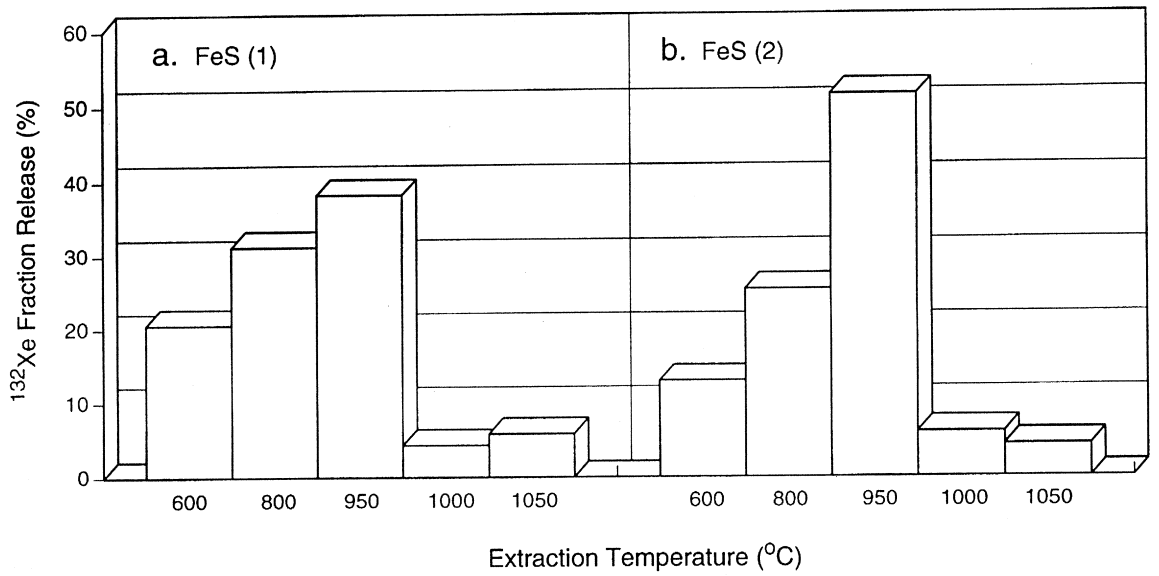


*Fig. 1. Release patterns of $^{132}$Xe from natural (Fig. 1a) and irradiated (Fig. 1b) sulfide samples during step-wise heating. The greatest release of $^{132}$Xe occured at 950°C. Higher purity sulfides were selected for irradiation and show the more pronounced release of $^{132}$Xe at 950°C.*

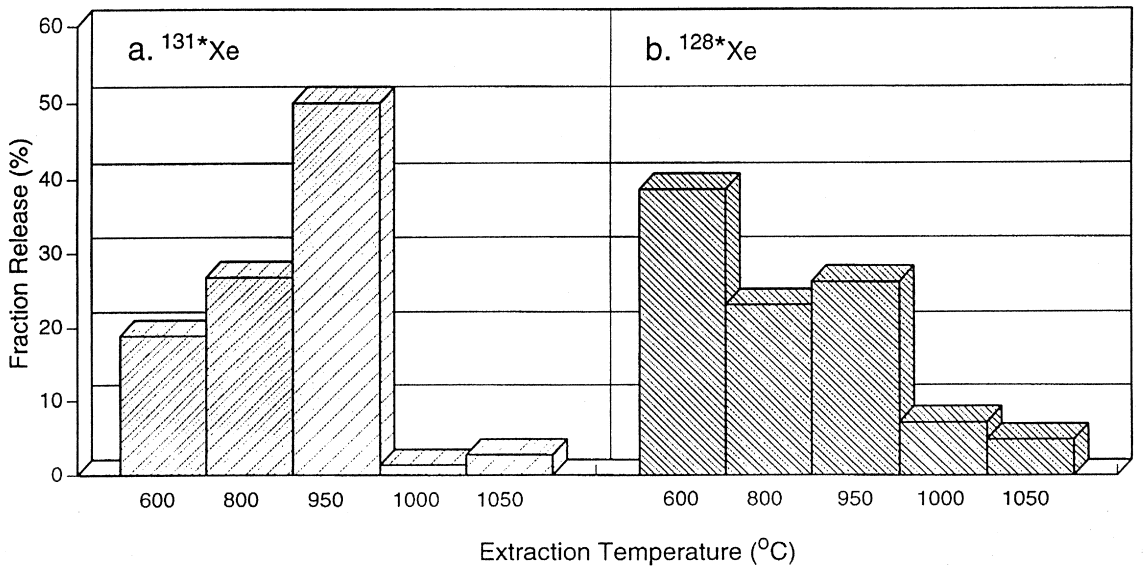


*Fig. 2. Release patterns of pile-produced $^{131*}$Xe (Fig. 2a) and $^{128*}$Xe (Fig. 2b) from the irradiated sulfide sample. The peak release of $^{131*}$Xe at 950°C indicates melting and degassing of the sulfides. Pile-produced $^{128*}$Xe is released over a broad temperature range because of the higher affinity of iodine than tellurium for the high temperature silicates.*

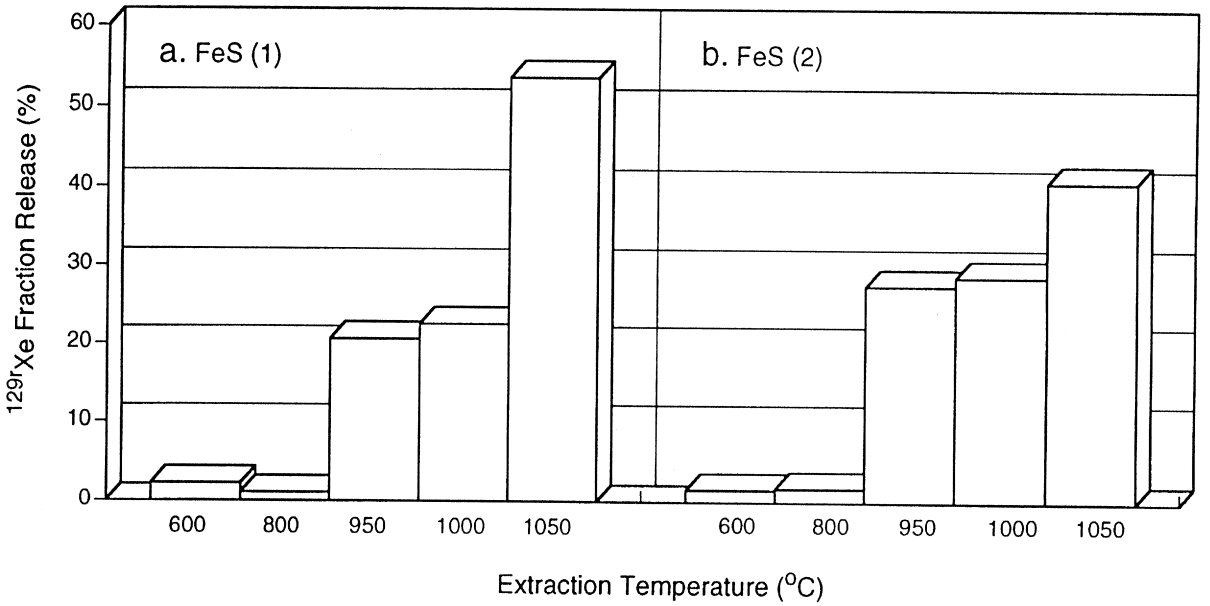


*Fig. 3. Release patterns of radiogenic $^{129}$Xe, $^{129r}$Xe, from natural (Fig. 3a) and irradiated (Fig. 3b) sulfide samples during step-wise heating. Both samples show the greatest release of $^{129r}$Xe at 1050°C. The more pronounced release of $^{129r}$Xe from FeS(1) at 1050°C (Fig. 3a) is attributed to a higher content of silicates in the unirradiated sample.*

same manner as shown in Fig. 1 for the release of $^{132}$Xe.

The release pattern of $^{131}$*Xe from FeS(2) (Fig. 2a) is very similar to that of $^{132}$Xe from FeS(2) (Fig. 1b), with peak release occurring at 950°C. Since the target element used to generate the $^{131}$*Xe is a chalcophile element, Te, the peak release of $^{131}$*Xe at 950°C indicates melting and degassing of the sulfides at this temperature.

A different release pattern was observed for $^{128}$*Xe, with peak release at 600°C. Differences in the release patterns of $^{131}$*Xe and $^{128}$*Xe are expected, since tellurium and iodine have different geochemical properties and therefore occupy different sites. This peak release of $^{128}$*Xe in the lowest temperature fraction has been observed from other irradiated meteorite samples (e.g., Merrihue, 1966; Niemeyer, 1979; Swindle *et al.*, 1988). The fraction of $^{128}$*Xe that remains in FeS(2) above 950°C, when the sulfides melt, is significantly higher than that of $^{131}$*Xe. We attribute this to the selective enrichment of iodine over tellurium in silicates, which start to degas above 950°C and release radiogenic $^{129}$Xe.

This can be seen by looking at the release of radiogenic $^{129}$Xe, $^{129r}$Xe, generated by the decay of extinct $^{129}$I in these samples. Figure 3 shows the release patterns for $^{129r}$Xe, calculated by assuming that the decay of 16 my $^{129}$I is responsible for the observation of $^{129}$Xe/$^{132}$Xe ratios higher than that in air. It can be seen in Fig. 3 that the bulk of the $^{129r}$Xe is released at the highest extraction temperature.

The total amount of $^{129r}$Xe in FeS(2) is $3.2 \times 10^{-10}$ ccSTP/g; in FeS(1) it is $5.1 \times 10^{-10}$ ccSTP/g. The release of $^{129r}$Xe from FeS(1) at 1050°C (Fig. 3a) is more pronounced than that from FeS(2) at 1050°C (Fig. 3b). We attribute both of these observations to the lithophillic nature of iodine and to a higher content of silicates in FeS(1) than in FeS(2).

*Xenon isotope ratios as indicators of nucleogenetic components*

The primary objective of this study was to identify the isotopic composition of xenon in FeS

from Allende, the primitive unequilibrated C3V carbonaceous chondrite in which nucleogenetic isotopic anomalies were first found for several elements (see, for example, reviews by Begemann, 1980, 1993; Lavrukhina, 1980; Clayton, 1981; Lee, 1988). Isotopes of these elements are heterogeneously distributed among different mineral phases of Allende, on a scale of several Å to a few mm in size.

Previous studies revealed a spike of terrestrial-type Xe released near the melting point of meteoritic FeS (Lewis *et al.*, 1979; Niemeyer, 1979; Hwaung and Manuel, 1982). Lewis *et al.* (1979) attribute the Xe-T to "absorbed atmospheric Xe". Niemeyer (1979) does not explain why the Xe released from Mundrabilla troilite is like Xe-T. Hwaung and Manuel (1982) claim Xe-T is a trapped component "that was dominant in the central Fe- and S-rich region of the protoplanetary nebula". To identify the extraction temperature at which Xe was released from FeS, half of our sample was irradiated to produce the tracer isotope, $^{131}$*Xe, in the sulfides.

The isotopic compositions of Xe released at each temperature from Allende's sulfides are shown in Table 1. As noted earlier, a peak release of the tracer isotope, $^{131}$*Xe, occurred at 950°C (Fig. 2a). Therefore, this is the temperature at which trapped Xe is released from these sulfides.

Xe released at 950°C from unirradiated FeS(1) and irradiated FeS(2) is shown in Fig. 4a by open and darkened symbols, respectively, with background lines for AVCC and terrestrial Xe. In this figure, the abundance of each stable isotope of mass, $i$, is defined by the ratio, $R^i$, where

$$R^i = \left({}^i\mathrm{Xe}/{}^{132}\mathrm{Xe}\right)/\left({}^i\mathrm{Xe}/{}^{132}\mathrm{Xe}\right)_{\mathrm{AVCC}}. \quad (1)$$

The presence of radiogenic $^{129}$Xe in meteoritic and terrestrial material precludes the use of $R^{129}$ in Fig. 4.

It can be seen from Fig. 4a that the isotopic ratios of Xe released from FeS(1) and FeS(2) are closer to Xe-T than to AVCC Xe, except for a small excess of spallogenic $^{126}$Xe in both samples, a slight deficiency of $^{130}$Xe in FeS(1), and excesses of pile-produced $^{128}$*Xe and $^{131}$*Xe in FeS(2).

From the results shown in Fig. 4a, we conclude that Xe-T, rather than AVCC-Xe, was initially trapped in Allende's FeS. Since Xe-T is also dominant in the Earth's atmosphere, it seems unlikely that this represents yet another example of interstellar, presolar material imbedded in this meteorite. A more plausible interpretation is that offered by Niemeyer (1979) to explain the I-Xe age of troilite in Mundrabilla, an IAB meteorite: "... the troilite formed in a different nebular region ... and at the time of agglomeration of the IAB meteorite, the troilite was transported to the IAB region and mechanically mixed with these other phases".

Likewise, we interpret the presence of Xe-T in Allende's FeS as evidence that this formed in the central nebular region where the Earth acquired the bulk of its Xe, Fe and S. Xenon in this region was predominately Xe-T. Some of the FeS which formed here was transported to the C3V region and agglomerated with other phases of Allende.

Other bodies that formed in the central region of the solar system also contain Xe-T. The darkened symbols in Fig. 4b are estimates of Xe in the martian atmosphere, based on analyses of meteorite EETA 79001. Becker and Pepin (1984) note that "The trapped xenon composition is consistent with addition of neutron-capture, radiogenic and fissiogenic isotopes to a base composition resembling terrestrial atmospheric Xe". The darkened circles represent this xenon. The darkened triangles are xenon that Swindle *et al.* (1986) calculated for SPB (Shergottite Parent Body) from high-temperature extractions of meteorite EETA 79001. They note that, "Normalized to $^{132}$Xe, the light isotope ratios are indistinguishable from air—and $^{134}$Xe and $^{136}$Xe are enhanced relative to the terrestrial atmosphere or AVCC". The martin xenon represented by both sets of darkened symbols is closer to Xe-T than to AVCC-Xe, but seems to contain excess $^{134}$Xe and $^{136}$Xe. However, the open squares represent xenon released from meteorite ALH 84001, which Swindle *et al.* (1995) suggest

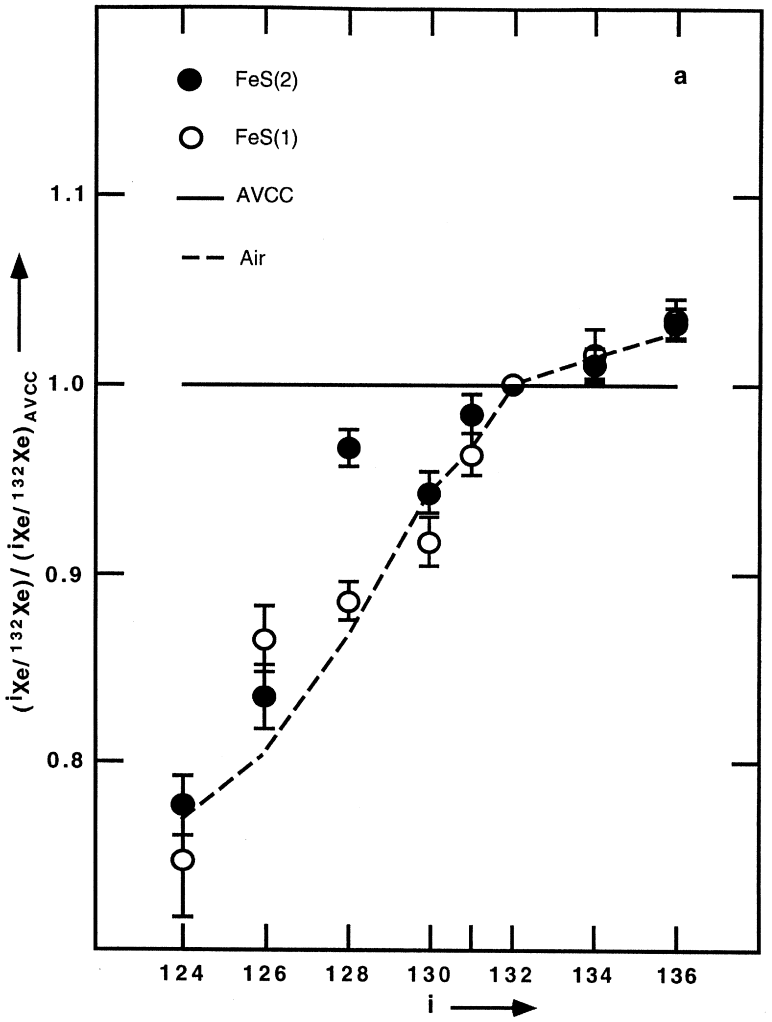


*Fig. 4. A comparison of air and AVCC xenon with that released at 950°C from Allende's FeS(1) and FeS(2) (Fig. 4a) and from meteorites (Becker and Pepin, 1984; Swindle et al., 1986, 1995) which trapped xenon from the martian atmosphere (Fig. 4b). The dashed line represents xenon in air; the solid line shows AVCC Xe. Values are not shown for $^{129}$Xe because of interference from radiogenic $^{129}$Xe. In Fig. 4a, xenon released when the Allende sulfides melt is closer to air than to AVCC xenon. In Fig. 4b, calculated abundances of trapped martian Xe isotopes in EETA 79001 are closer to air than to AVCC Xe, but contain excess $^{134,136}$Xe (Becker and Pepin, 1984; Swindle et al., 1986). Excess $^{134,136}$Xe is not observed in ALH 84001 (Swindle et al., 1995), and the light isotopes are not corrected for spallation, most notable as excess $^{126}$Xe. The trapped martian xenon in ALH 84001 is also generally closer to air than to AVCC Xe.*

"has incorporated a relatively large amount of martian atmosphere". This has not been corrected for spallation. If corrected, relative abundances of the light Xe isotopes in ALH 84001 would become more like Xe-T and the Xe seen in EETA 79001, but $^{134}$Xe and $^{136}$Xe are not enriched in ALH 84001. The data in Fig. 4b suggest that the base composition of xenon in the martian atmosphere is closer to Xe-T than to AVCC Xe.

Xe-T also appears to be the main xenon component in the sun. Mass fractionation of Xe-T, to enrich the lighter isotopes by about 4% per mass unit, gives a close, but not perfect, match to the isotopic composition of solar-wind implanted xe-

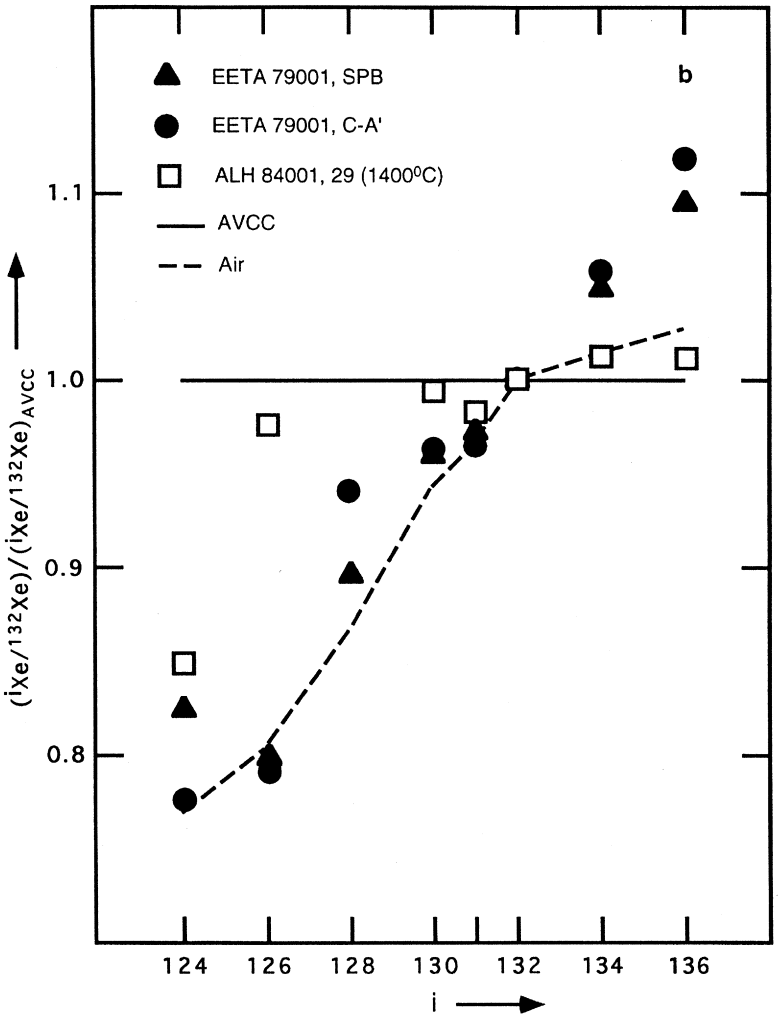


*Fig. 4. (continued).*

non. This can be seen in Fig. 2 of Boulos and Manuel (1971). Fractionated AVCC Xe does not match the composition of solar xenon nearly as well as fractionated Xe-T, as can be seen in Figs. 5 and 6 of Kaiser (1972) or Fig. 3 by Bernatowicz and Podosek (1978).

Thus, it appears that Xe-T is a major component of xenon in the Sun, in FeS mineral separates of diverse meteorites, and in the atmospheres of two iron-rich planets, Earth and Mars. Atmospheric contamination is not a likely source of Xe-T in these extra-terrestrial samples for the following reasons:

a) The peak release of Xe-T from meteorites EETA 79001 and ALH 84001 (Fig. 4b) occurs at very high temperatures (1400°–1500°C), long after surface adsorbed Xe has been removed.

b) The peak release of Xe-T from FeS of diverse meteorites occurs at 900°–1100°C. It is unlikely that this represents surface adsorbed Xe from air because the same high-vacuum, step-wise gas extraction techniques have been successfully employed in this laboratory to quantitatively determine ultra-trace levels of radiogenic $^{128}$Xe and $^{130}$Xe that accumulated from $\beta\beta$-decay of $^{128}$Te and $^{130}$Te in Te-minerals that melted and released

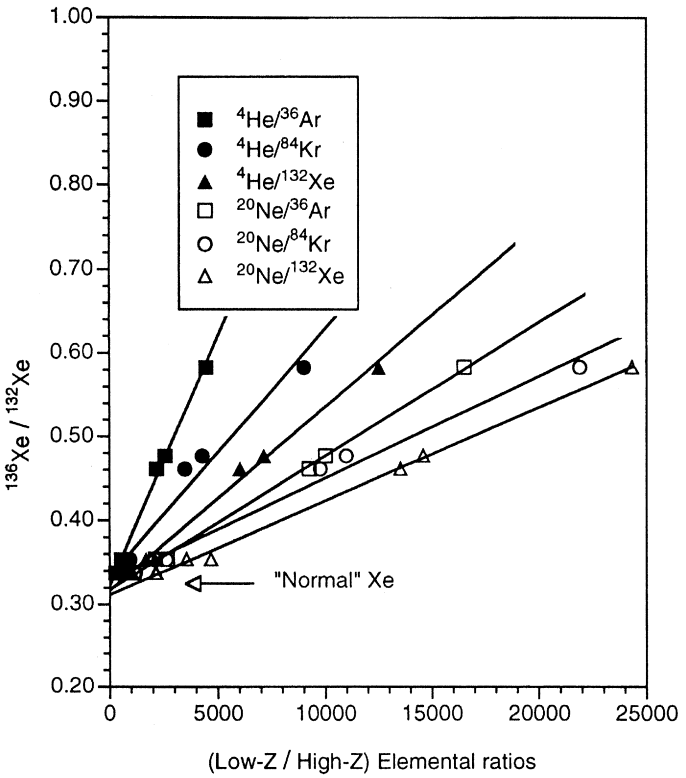


*Fig. 5. Correlations of isotopic ratios of a high-Z noble gas, Xe, with relative abundances of the low-Z noble gases, He and Ne. Relative values of six elemental ratios, $^{4}He/^{36}Ar \times 5$, $^{4}He/^{84}Kr \times 0.08$, $^{4}He/^{132}Xe \times 0.15$, $^{20}Ne/^{36}Ar \times 3800$, $^{20}Ne/^{84}Kr \times 40$, $^{20}Ne/^{132}Xe \times 60$, are shown on the abscissa. This figure shows how these six elemental ratios correlate with values of the $^{136}Xe/^{132}Xe$ isotopic ratio. The correlations converge on the values of isotopically "normal" Xe at $^{4}He = {}^{20}Ne = 0$. Data, from Lewis et al. (1975), display such correlations for isotope ratios of Xe, Kr and Ar.*

Xe at even lower temperatures (e.g., Lee *et al.*, 1991). It is also unlikely that Xe-T is released from reaction of molten FeS with the crucible because Hwaung and Manuel (1982) used the same type of stainless steel tube for gas extraction of FeS from iron meteorites. On melting, the amount of Xe released per gram of sample was almost three order-of-magnitude less than that observed from Allende's FeS.

*Correlated chemical and isotopic heterogeneities*

Stellar nuclear reactions generate different isotopes of a heavy, high-Z element like Xe at different stellar depths, at different stages of stellar evolution and by different nuclear reactions, e.g., the p-, r- and s-processes of nucleosynthesis (Burbidge *et al.*, 1957). An evolved star also becomes chemically layered with light, low-Z elements like H and He existing only in the surface layers, and more stable nuclear species like Fe-Ni enriched near the core because "nuclear evolution is most advanced in the central region and least or not at all advanced near the surface" (Burbidge *et al.*, 1957). Accordingly, fresh stellar debris may retain chemical and isotopic heterogeneities if the supernova explosion did not result in complete mixing.

In addition to Xe-T, other examples of isotopically anomalous xenon trapped in Allende and other primitive carbonaceous chondrites are Xe-S (Srinivasan and Anders, 1978) and Xe-X (or Xe-HL) (Manuel *et al.*, 1972). Summarized below are

results which indicate that chemical heterogeneities from nucleosynthesis also survived and were interlinked with the isotopic inhomogeneities observed in xenon and other elements:

i) Lewis *et al.* (1975) first isolated mineral fractions of Allende that contained Xe-X. In these separates, isotopic ratios of the high-Z noble gases, Ar, Kr and Xe, correlate linearly with relative abundances of the low-Z noble gases, He and Ne (Manuel and Sabu, 1975, 1977). An example of these correlations is shown in Fig. 5 as " best fit" lines between $^{136}Xe/^{132}Xe$ and low-Z/high-Z elemental ratios—$^{4}He/^{36}Ar$, $^{4}He/^{84}Kr$, $^{4}He/^{132}Xe$, $^{20}Ne/^{36}Ar$, $^{20}Ne/^{84}Kr$ and $^{20}Ne/^{132}Xe$. These six correlation lines tend to converge on a single value for the isotopic ratio of the high-Z element as the abundances of the low-Z elements extrapolate to zero. Similar correlations are observed for the noble gases in mineral separates of other meteorites, and in each case the value of the isotopic ratio of the high-Z element at the intercept is approximately that of the "normal" Ar, Kr and Xe found in bulk meteorites and the inner planets (Ballad *et al.*, 1978; Sabu and Manuel, 1980).

ii) Clayton *et al.* (1976) showed that the level of excess $^{16}O$ in different types of meteorites correlates with their geochemical classification. On the basis of excess $^{16}O$, they also identified six categories of meteorites, with the Earth and its moon being part of the terrestrial group that includes enstatite chondrites and some differentiated meteorites. Chemical heterogeneities that accompanied isotopic inhomogeneities in the protoplanetary nebula offer a direct explanation for the correlation of $^{16}O$ with the geochemical classification of meteorites.

iii) Even prior to this study, the interlinkage of Xe-X with diamond and of Xe-S with silicon carbide was so strong that Huss and Lewis (1995) were able to use isotopic measurements on Xe and other noble gases in acid residues of seven classes of chondrites to estimate the abundances of diamonds and silicon carbide in these meteorites. Clearly, isotopic measurements could not be used to calculate the amounts of chemically distinct carrier phases unless the isotopic and chemical properties were closely connected. The scale of these interlinked chemical and isotopic heterogeneities is reflected in their conclusion that all chondrite classes sampled "the same solar-system-wide reservoir" of carrier grains (Huss and Lewis, 1995). Likewise, we note that the interlinkage of Xe-T with FeS may extend from the terrestrial planets (Becker and Pepin, 1984; Swindle *et al.*, 1986, 1995) to iron meteorites (Niemeyer, 1979; Hwaung and Manuel, 1982) to the Allende carbonaceous chondrite.

iv) With the results of this study, three isotopically distinct forms of Xe have been identified in three chemically distinct host minerals that may indicate variations in the depth of stellar nucleosynthesis: Xe-X, from the outer stellar layers, is trapped with large amounts of the He and Ne (Fig. 5) in carrier grains (diamond) of element 6. Xe-S, from an intermediate stellar region where products of slow neutron capture during stellar evolution survived the subsequent supernova event, is trapped in SiC, with an average atomic number of 10. Xe-T, from the stellar interior where iron group elements were produced (Burbidge *et al.*, 1957), is trapped in FeS with an average atomic number of 21.

v) Recent ion probe measurements of Be and B in chondrules reveal correlations between isotopic enrichments of $^{11}B$ and values of the B/Si and Be/Si elemental ratios (Chaussidon and Robert, 1995). These interlinked chemical and isotopic heterogeneities, though less obvious than those shown in Fig. 5 for meteoritic noble gases, were interpreted as evidence for the nucleosynthesis of $^{11}B$-rich boron in the pre-solar cloud.

The five items cited above indicate chemical and isotopic inhomogeneities from nucleosynthesis were interlinked over vast regions of the protosolar nebula.

## CONCLUSIONS

The results of this study show that Allende's FeS is the carrier of Xe-T. This provides additional evidence of local element synthesis (Manuel and Sabu, 1975, 1977) and interlinked isotopic and

chemical heterogeneities in material that formed the solar system. It also confirms a few earlier studies showing a possible association between Xe-T and FeS in mineral separates of diverse meteorites—the Allende C3V carbonaceous chondrite (Lewis *et al.*, 1979), the Mundrabilla IAB iron meteorite (Niemeyer, 1979), the Cape York IIIA, and the Odessa IA iron meteorites (Hwaung and Manuel, 1982).

The dominance of Xe-T in the atmospheres of Earth (and Mars) and in meteoritic FeS suggest a radially heterogeneous protoplanetary nebula in which Xe-T was initially associated with the Fe- and S-rich region where the terrestrial planets formed. Likewise, the dominance of Xe-X in meteoritic diamond and the high abundances of He and Ne in this noble gas component (see Fig. 5) may indicate its initial association with the outer portion of the protoplanetary nebula where the giant Jovian planets formed from material rich in low-Z elements.

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This paper is dedicated to the memory of another former student of Professor Kuroda, Dr. Dwarka Das Sabu, an unusually talented scientist who accepted and dealt with the pain and certainty of death from cancer in 1991 with the same tranquility and detachment that he displayed earlier in interactions with the scientific community.

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