

## SEARCH FOR ISOTOPIC ANOMALIES CORRELATED WITH CCF XENON.

## I. BARIUM

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**Introduction.** CCF ("carbonaceous chondrite fission") xenon is one of the oldest, largest, and yet least understood isotopic anomalies in meteorites. The enrichment in the heavy isotopes 131-136 has been attributed either to *in situ* fission of a long-lived ( $>10^6$  yr) superheavy element in the early solar system [1] or to *direct nucleosynthesis*: *r*-process [2], *n*-process [3], or fission of short lived ( $<1$  d) superheavy elements made in the *r*-process [4]. The concurrent enrichment of the light isotopes 124-130 may then be attributed either to mass fractionation during trapping [5,6] or to some variant of the *r*-process [2,7]. Both models have their problems, however. The fission hypothesis requires the fissioning element to be surrounded by  $\sim 10\mu\text{m}$  carbon clusters, for efficient trapping of fission fragments. It also requires close, though not perfect coupling of the light and heavy Xe anomalies. The supernova hypothesis, on the other hand, must explain the absence of a <sup>129</sup>Xe anomaly [8], the isotopic normalcy of the carbonaceous carrier of CCFXe [9], and the actual isotopic patterns of Kr and Xe, which seem to require lower temperatures and fluxes than the *r*-process [3].

An obvious test of both models would be detection of isotopic anomalies in neighboring elements, especially Ba and the light REE. With additional data points in the mass range 130-154, the pattern may be sufficiently well defined for a conclusive match or mismatch. However, the expected anomalies are on the order of  $10^{11}$  atoms/g of carrier, and, hence, require samples of very low Ba and REE content as well as careful chemistry and mass spectrometry.

**Samples.** We analyzed a 152.0 mg sample of Allende residue CD, whose genealogy was as follows: Allende bulk (HF, HCl)  $\rightarrow$  A12 (0.573 wt%) + All.13 (0.035%). A12  $\equiv$  CA (colloid separation)  $\rightarrow$  CB (0.2367%; HCl + HF and HCl)  $\rightarrow$  CD (0.2228%) and solutions CD1, CD2. Sample CD was then treated with a succession of high purity acids that dissolved phases Q, gas-poor carbon (C<sub>g</sub>), and resistant carbon (C<sub>r</sub>) and chromite (Table 1). Noble-gas data for this series are not yet available, and we are, therefore, listing data for an analogous series of samples (BA, BB, BG and BT; [9]).

**Results.** Data for Ba are shown in Table 2 as relative deviations from the normal; those for Nd, Sm and Sr are given in the companion abstract by Lugmair *et al.* [10]. Ba was extracted from the same unspiked and spiked aliquots ( $\sim 10\%$  of total solution) from which the other elements were separated. Mass spectrometric procedures were developed which yield high precision data for the major isotopes for samples as small as 1 ng. The normal isotopic ratios were determined from a standard solution made from high purity BaCO<sub>3</sub>. As a control sample, we used a terrestrial tholeiite (E-2a); the Ba data are given in Table 2. The isotopic ratios were corrected for mass fractionation to <sup>135</sup>Ba/<sup>138</sup>Ba = 0.091940 [11]. As is evident from the data in Table 2, no deviations from normal isotopic ratios do exist in any of the etch fractions. But, for a comparison of the Ba data with CCFXe, <sup>135</sup>Ba is especially important. Accordingly, the data were re-normalized to <sup>134</sup>Ba/<sup>136</sup>Ba, the two major isotopes of Ba which are shielded from  $\beta^-$ -decay. This choice of isotopes leads to a marked increase in uncertainties but allows extreme upper limits for excesses in <sup>135</sup>Ba to be determined. These are given in the last column of Table 2 and may be compared with the values in Table 1.

**Discussion.** The most striking and significant result is the absence of a detectable <sup>135</sup>Ba anomaly. The extreme upper limits on such an anomaly— $\sim 10^{10}$  atoms/g of CD (Table 2)—are about 1 order of magnitude smaller than the <sup>136</sup>Xe anomaly in the fractions with the largest concentrations of Xe<sub>f</sub> (Table 1)! This is a major paradox for all models—fission or neutron capture—as any process that makes <sup>134,136</sup>Xe must make comparable amounts of <sup>135</sup>Ba. Given the volatility of Xe, one would expect non-volatile elements to have larger, not smaller absolute anomalies!

The magnitude of the problem is illustrated in Figures 1 and 2, showing the predictions of the *in situ* fission and nucleosynthesis models together with *extreme* upper limits for possible excesses on the isotopes measured. Three predicted patterns are shown: fission of the long-lived doubly magic nuclide 114<sup>298</sup> [4], fission of a mix of short-lived nuclides of  $A \approx 280$  made in the *r*-process [4,12], and the solar-system *r*-process distribution [13] (recalculated for abundances from [14]). (The conventional *r*-process does not account for the isotopic composition of CCFXe, but this defect can be remedied by the slower and cooler "*n*-process" [3] which shifts the peak closer to the line of  $\beta$ -stability. For the purpose of the present paper, this difference is not significant.)

None of the three patterns predicts such low enrichments for the unshielded Ba isotopes (or for Nd and Sm isotopes; see [10], this volume). We can at once dismiss the trivial explanation that anomalous Ba, Nd and Sm were leached out of Allende CD during the month-long treatment with HCl and HF. CCFXe, as well as two Nd anomalies, have survived this treatment [10], and so it is unlikely that other anomalies for Ba, Nd and Sm were depleted by 2 orders of magnitude during this treatment.

For the nucleosynthesis models, chemistry offers a way out of this dilemma. Ba, Nd and Sm all have high condensation temperatures but low first ionization potentials, compared to Xe. Accordingly, these three elements may have been separated from Xe by prior condensation or by plasma processes in the expanding supernova shell [15].

No such excuse is available for the *in situ* fission model, as all fission products ought to be trapped in the carbon "catcher" with equal efficiency. Though a tiny loophole remains (fissionogenic Ba may have been leached by HCl-HF, even though anomalous <sup>142,143</sup>Nd wasn't [10]), the present data seem to rule out the *in situ* fission model.

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References: [1] Anders *et al.* (1975), *Science* 190; [2] Manuel *et al.* (1972), *Nature* 240; [3] Blake and Schramm (1976), *Nature* 263; [4] Steinberg and Wilkins (1978), *Astrophys. J.* 223; [5] Lewis *et al.* (1975), *Science* 190; [6] Lewis *et al.* (1977), *J. Geophys. Res.*, 82; [7] Srinivasan (1981), *Naturwissenschaften.*, 68; [8] Lewis and Anders (1981) *Astrophys. J.* 247; [9] Swart *et al.* (1982), *Science*, in press; [10] Lugmair *et al.* (1982) this volume; [11] Eugster *et al.* (1969), *J. Geophys. Res.* 74; [12] Blake and Schramm (1974), *Astrophys. Space Sci.* 30; [13] Käppeler *et al.* (1982), *Astrophys. J.* 257; [14] Anders and Ebihara (1982), *GCA* 46; [15] Arrhenius and Alfvén (1971), *EPSL* 10.

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Table 1:

Sample or solution <sup>a)</sup>	wt %	Solvent, T(°C)	Phases diss.	<sup>132</sup> Xe (10 <sup>-8</sup> cc/g)	<sup>136</sup> Xe/ <sup>132</sup> Xe	<sup>136</sup> Xe <sub>f</sub> (x 10 <sup>11</sup> at/g CD)	<sup>135</sup> Ba <sub>ex</sub> <sup>c)</sup> (x 10 <sup>11</sup> at/g CD)
CD	≈ 100 <sup>b)</sup>	HNO <sub>3</sub> , 75°		60.7	0.3403	4.94	
<u>CE-1</u>			<u>Q</u>			<u>1.14</u>	0.03
CE	97.8	HClO <sub>4</sub> , 140°		9.9	0.464	3.80	
<u>CF-1</u>			<u>C6</u>			<u>0.21</u>	0.13
CF	28.4	HClO <sub>4</sub> , 200°		19.4	0.641	3.58	
<u>CG-1</u>			<u>Cy, Chr</u>			<u>3.58</u>	0.20
CG	9.8			(=0)		(=0)	

a) Solution labels are shown underlined.  
 b) This sample represents 0.2228 weight % of its bulk Allende parent.  
 c) Upper limit of excess <sup>135</sup>Ba with standard normalization.

Table 2: Ba isotopic variations - All. CD (in parts in 10<sup>4</sup>)<sup>a)</sup>.

Fraction	ε(137/138)	ε(136/138)	ε(134/138)	ε(132/138)	ε(130/138)	[ <sup>135</sup> Ba <sub>ex.</sub> ] <sup>b)</sup> x 10 <sup>11</sup> at/g CD
CE-1	- 1.0 ± 1.0	+ 0.5 ± 1.3	+ 0.6 ± 2.4	+ 5.7 ± 21.2	+ 11.5 ± 27.1	< 0.14
CF-1	- 0.3 ± 0.3	+ 0.5 ± 0.5	+ 1.2 ± 1.2	- 1.4 ± 14.2	+ 4.7 ± 13.6	< 0.20
CG-1	- 0.5 ± 0.9	+ 0.5 ± 1.3	+ 1.2 ± 1.5	+ 17.0 ± 15.5	- 2.0 ± 13.6	< 0.40
Terr. Thol.						
E-2a (10 ng)	- 0.3 ± 0.3	- 0.3 ± 0.5	- 0.3 ± 1.2	+ 2.8 ± 7.1	+ 11.5 ± 6.8	

a) Corr. for fractionation to <sup>135</sup>Ba/<sup>138</sup>Ba = .09194<sup>[11]</sup>; the normal ratios R(i). (rel. to <sup>138</sup>Ba) are: R(137) = .156545 ± 3, R(136) = .109543 ± 4, R(134) = .033716 ± 3, R(132) = .001413 ± 1, R(130) = .001475 ± 1.  
 b) Extreme upper limit for excess <sup>135</sup>Ba.

