BARIUM AND NEODYMIUM ISOTOPIC ANOMALIES IN THE ALLENDE METEORITE

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ABSTRACT

Isotopic anomalies have been found for Ba and Nd in two inclusions from the Allende meteorite. These inclusions are typical Ca-Al-rich objects associated with early condensates from the solar nebula but have distinctive O and Mg isotopic anomalies of the FUN type. Sample C1 shows a depletion only in ¹³⁵Ba of 2 parts in 10⁴ and normal Nd. Sample EK1-4-1 shows large positive excesses in the unshielded isotopes ¹³⁵Ba and ¹³⁷Ba of 13.4 and 12.3 parts in 10⁴, respectively. The Nd isotopic composition in EK1-4-1 is highly aberrant in at least five isotopes. Both the Ba and Nd anomalies can be explained by a model of *r*-process addition. These anomalies are found to be uniform between coexisting mineral phases. These observations show the existence of substantial isotopic anomalies in refractory elements in the neighborhood of Xe and extend the range of elements showing isotopic effects to O, Ne, Mg, Ca, Kr, Xe, Ba, and Nd. The observations, in conjunction with the presence of ²⁶Al ($\tau_{1/2} = 7 \times 10^5$ years), are interpreted as the result of a nearby supernova explosion which produced elements over a wide mass range and injected them into the early solar nebula shortly before condensation.

Subject headings: meteors and meteorites — nucleosynthesis — stars: supernovae

I. INTRODUCTION

Ba and Nd isotopic anomalies have been discovered in one inclusion in the Allende meteorite and Ba isotopic anomalies in a second inclusion from the same meteorite. Anomalies in Ca from the same inclusions are reported in a companion *Letter* by Lee, Papanastassiou, and Wasserburg (1978). A new search for isotopic variations in Ba was undertaken because of the existence of isotopic anomalies in O and Mg in Allende inclusions. Ba has seven stable isotopes which are believed to have been made in the p-, s-, and r-processes (Burbidge et al. 1957, hereafter B2FH). The relative contributions to each of the isotopes by these processes are quite different. The existence of isotopic anomalies in meteoritic, terrestrial, and solar Xe which are not attributable to the radioactive decay of known nuclides also suggested that nuclear effects might be present in the neighborhood of atomic number $\breve{Z} = 56$. Previous studies of Ba using high-precision methods were re-ported by Eugster, Tera, and Wasserburg (1969, hereafter ETW), who found no evidence for nonlinear isotopic shifts to a level of 10^{-4} and a maximum mass fractionation of 2×10^{-4} per mass unit. Measurements of ¹⁴²Nd/¹⁴⁴Nd from an inclusion from Allende have been published by Scheinin, Lugmair, and Marti (1976). These workers have tentatively inferred small excesses in ¹⁴²Nd which they have attributed to decay of extinct ¹⁴⁶Sm. However, the inherent precision of the data was not sufficient to clearly resolve this effect.

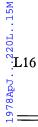
Inclusions in which Ba and Nd isotopic effects were found are Ca-Al-rich objects typical of the type considered as high-temperature condensation products in the solar nebula (see Grossman 1972). They contain

* Division Contribution No. 2979 (252).

coarse-grained fassaitic pyroxene, melilite, spinel, and plagioclase. They also contain fine-grained alteration products associated with melilite and plagioclase which are rich in a "sodalite"-like material and believed to result from a late-stage reaction of early condensates with cooler, volatile-rich material in the solar nebula which altered the oxygen isotopic composition of the melilite (Clayton and Mayeda 1977; Wasserburg, Lee, and Papanastassiou 1977, hereafter WLP). These inclusions are known to be distinctive, as they contain magnesium with negative values of δ^{26} Mg and oxygen which does not lie on the usual correlation line between "normal" oxygen (O_N) and extraordinary oxygen (O_E) consisting of nearly pure ¹⁶O (Clayton, Grossman, and Mayeda 1973). The Mg in these inclusions shows $^{26}\mathrm{Mg}/^{24}\mathrm{Mg}$ ratios shifted from normal values by 2% and 3% per mass unit, favoring the heavier isotopes. These large shifts are almost quantitatively what would be expected by mass fractionation (F) with small but distinct residual negative anomalies in ²⁶Mg which are attributed to unknown nuclear processes (UN). This class of anomalies was designated as FUN anomalies (WLP).

II. RESULTS

Procedures were similar to those described by ETW. Data were taken on the Lunatic I and III mass spectrometers (Wasserburg *et al.* 1969) which give the same self-consistent Ba isotopic ratios within 3 parts in 10⁵. An improvement in sensitivity by a factor of 50 and in precision by a factor of 2 was achieved over ETW. Typical ion beams for ¹³⁸Ba were 2×10^{-11} A when collecting data on masses 134–138. Data for the rare isotopes 130–132 were taken with the ¹³⁸Ba ion beam at 10^{-10} A. Data are given in Table 1 for isotope k as



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TABLE 1

BARIUM ISOTOPIC VARIATIONS*

Sample [†]	Ba(ppm)	€137	<i>€</i> 136 Normalized to	<i>€</i> 134 • ^{1 3 5} Ba/ ^{1 3 8} Ba; Index	€132 Isotope ¹³⁸ Ba	€130
EK1-4-1 SC EK1-4-1 SC EK1-4-1 MEI	b‡	$+7.9\pm0.4$ +7.8±0.5 +8.2±0.5	-9.7 ± 0.6 -9.5 ± 0.6 -8.7 ± 0.6	-17.8 ± 1.2 -17.2 ± 1.8 -16.0 ± 2.4	-28.0 ± 14.0 -21.0 ± 7.0 -28.0 ± 14.0	-34.0 ± 7.0 -41.0±14.0 -41.0±7.0
C1 a [‡] § C1 b [‡] C1 c [‡] C1 S2 C1 PYX	39 17	$+0.9\pm0.4$ +0.1±0.4 +0.2±0.4 -0.1±0.4 +0.4±0.4	$+1.9\pm0.6$ +1.5±0.6 +1.5±0.5 +1.5±0.6 +1.8±0.7	$\begin{array}{c} +2.1 \pm 1.5 \\ +2.1 \pm 0.9 \\ +3.0 \pm 1.2 \\ +3.0 \pm 1.5 \\ +3.9 \pm 1.5 \end{array}$	$\begin{array}{c} 0.0 \pm 7.0 \\ 0.0 \pm 7.0 \\ + 7.0 \pm 7.0 \\ + 3.0 \pm 14.0 \\ 0.0 \pm 7.0 \end{array}$	$\begin{array}{c} -10.0 \pm 20.0 \\ +10.0 \pm 7.0 \\ +10.0 \pm 7.0 \\ +14.0 \pm 14.0 \\ +7.0 \pm 7.0 \end{array}$
Normals		0.0 ± 0.4 -0.2±0.4	-0.1 ± 0.5 0.0 ± 0.6	-0.6 ± 0.9 +0.6±1.2	0.0±7.0 0.0±7.0	+3.0±7.0 +3.0±7.0
Enriched Stand (€137=+3.8, € (€137=+7.9, €1	134=+3.6)	+3.5±0.5 +8.0±0.6	$+0.3\pm0.6$ -0.2 ± 0.6	+5.0±1.2 +7.4±1.5		
		€137	€136 Normalized t	€135 :o ^{1 3 4} Ba/ ^{1 3 8} Ba; Inde	€132 ex Isotope ¹³⁸ Ba	€130
EK1-4-1 C1		$+12.3\pm0.4$ -0.6±0.4	-0.8 ± 0.6 +0.6±0.6	$+13.4\pm1.0$ -1.8±0.7	-1.0 ± 14.0 -7.0 ±7.0	$+2.0\pm7.0$ -7.0±14

*Errors are $2\sigma_{\text{mean}}$. Deviations in parts per ten thousand relative to our grand mean of terrestrial normals: ${}^{137}Ba/{}^{138}Ba = 0.156515\pm4$; ${}^{136}Ba/{}^{138}Ba = 0.109540\pm4$; ${}^{134}Ba/{}^{138}Ba = 0.033710\pm4$; ${}^{132}Ba/{}^{138}Ba = 0.001412\pm1$; ${}^{130}Ba/{}^{138}Ba = 0.001476\pm1$; ${}^{135}Ba/{}^{138}Ba = 0.091940$. ${}^{\dagger}PYX = pyroxene$; MEL = melilite; S2, SC = splits of crushed inclusion. ${}^{\ddagger}Repeat$ analysis of solution. ${}^{\$}Analyzed$ on L III mass spectrometer.

deviations from the normal ratios in parts in 10⁴ with ¹³⁸Ba as the index isotope:

$$\epsilon k = (R_{k/138}^{\rm C}/R_{k/138}^{\rm N} - 1) \times 10^4 \tag{1}$$

where $R_{k/138}^{N} = ({}^{k}Ba/{}^{138}Ba)_{normal}$ and $R_{k/138}^{C} = ({}^{k}Ba/{}^{138}Ba)$ measured in the sample and normalized for mass fractionation assuming ${}^{135}Ba/{}^{138}Ba$ to equal the terrestrial value. Standards enriched in ${}^{137}Ba$ and ${}^{134}Ba$ were measured to establish the sensitivity to which nonlinear anomalies could be detected (see Table 1).

Samples from Allende, including Ca-Al-rich chondrules and aggregates, olivine chondrules, a pyroxene chondrule, and total meteorite, were analyzed for Ba. With the exception of the FUN samples, C1 and EK1-4-1, all other samples yield a Ba isotopic composition identical to terrestrial Ba within limits of error (see Fig. 1). A preliminary report on these data, excluding C1 and EK1-4-1, was published by McCulloch, Papanastassiou, and Wasserburg (1976). A small sample $(2 \times 10^{-8} \text{ g})$ of Ba from high-purity anorthite from WA, which shows a 10% excess of ²⁶Mg, and a large sample from the associated pyroxene, were analyzed and found to be normal. The first analysis of Ba from C1a showed possible effects in ¹³⁷Ba/¹³⁸Ba, ¹³⁶Ba/¹³⁸Ba, and ¹³⁴Ba/¹³⁸Ba. An aliquot of the solution was again separated and analyzed, and the anomalies in ¹³⁶Ba/¹³⁸Ba and ¹³⁴Ba/¹³⁸Ba were reproduced. The smaller anomaly in ¹³⁷Ba/¹³⁸Ba was not reproduced. A second split of the bulk inclusion (C1-S2) and a sample of pyroxene from C1 were analyzed and yielded the same shifts in ¹³⁶Ba/¹³⁸Ba and ¹³⁴Ba/¹³⁸Ba. The nonlinear anomalies, while small, are reproducible by using different spectrometers and different samples.

The only discrepancy is the measurement of ¹³⁷Ba/ ¹³⁸Ba for C1a. A sample of EK1-4-1 SC, consisting dominantly of pyroxene, was analyzed and yielded extremely large deviations which exceed the 2 σ_{mean} errors by over a factor of 10. A second mass spectrometer run was performed and isotopic ratios were determined over a wide range in filament temperature. Data were taken for masses 134-138 with the ion beam ranging from 0.5 to 2×10^{-11} A to test for changes in the ratios. The values for those isotopic ratios that could be measured at these emission levels were all identical within error to the means for the high-intensity data of both SC samples. Data for masses 130-135 were taken with the ion beam at 8×10^{-11} A and were also identical with those obtained in the previous run. In addition, a melilite separate from EK1-4-1 was analyzed and yielded an isotopic composition identical to that found for the SC sample. These data demonstrate large nonlinear isotopic anomalies in EK1-4-1 which are the same for different phases with Ba concentrations differing by 40%. For all runs the spectrum was scanned at high sensitivity between masses 120 and 160 and no interfering masses were identified. The only peaks seen were due to LaO and CeO, which were present at a level of 2×10^{-12} A or less and do not interfere with Ba. During data collection the ratios Ba⁺/LaO⁺ and Ba⁺/ CeO⁺ changed by over a factor of 2 without any changes in the Ba ratios.

Upon finding effects in Ba, we deemed it necessary to establish whether anomalies existed in other higher-Z elements, and a study of Nd was made. The procedures for Nd are similar to those described by DePaolo and Wasserburg (1976). Nd data are presented in No. 1, 1978

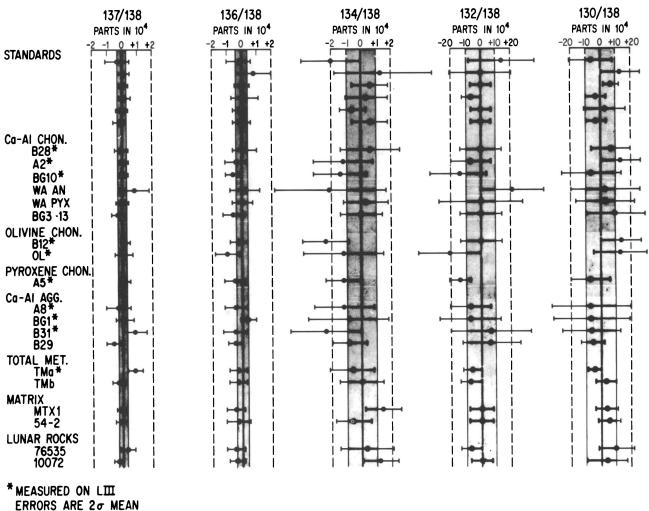
Table 2 as deviations (see eq. [1]) from standard normal ratios, with ¹⁴⁴Nd as the index isotope and mass fractionation corrections applied by using ¹⁵⁰Nd/¹⁴²Nd. Sample C1-S2 has a Nd isotopic composition indistinguishable from terrestrial normal. However, sample EK1-4-1 SC has large deviations of Nd isotopic composition from normal which exceed the errors by over a factor of 30. Data were collected for all isotopes with the ¹⁴⁴NdO⁺ beam ranging from 1 to 6×10^{-11} A, and isotopic ratios remained constant within experimental errors. The melilite separate from EK1-4-1 was analyzed for Nd and gave an isotopic composition identical to that of SC, except for mass 143, which is affected by ¹⁴⁷Sm decay. The Nd concentration in the melilite is lower than that in SC by a factor of 2.4. For the

samples C1 and EK1-4-1 the mass fractionation correction applied to both Ba and Nd was less than $\sim 3\%$ per mass unit and within the same range as applied to the terrestrial normal.

III. DISCUSSION

Most samples of the Allende meteorite contain Ba of normal isotopic composition. Some of the samples analyzed for Ba had substantial contributions of O_E (up to 3%) and of excess ²⁶Mg (up to 10%) correlated with ²⁷Al. It follows that no obvious correlation exists between these nuclear effects at low Z and Ba. In contrast, the two inclusions which are distinguished by having FUN anomalies also show distinct nonlinear isotopic anomalies in Ba. Inclusion C1 has anomalies at

BARIUM ISOTOPIC COMPOSITION NORMALIZED TO 135/138 = 0.09194



SHADED AREA IS 20 MEAN OF STANDARDS

FIG. 1.—Bar graph showing fractional deviations in the isotopic composition of Ba relative to normal for all Allende samples and two lunar samples studied in this work excluding FUN samples C1 and EK1-4-1. Reference line at 0 is mean of terrestrial values.

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TABLE 2 Neodymium Isotopic Variations*

Sample	Nd (ppm)	Sm (ppm)	€142	€143 [†]	€145	€146	<i>€</i> 148	€150			
		Normalized to ¹⁵⁰ Nd/ ¹⁴² Nd; Index Isotope ¹⁴⁴ Nd									
EK1-4-1 SC a EK1-4-1 SC b EK1-4-1 MEL	11.7 11.7 4.9	4.1 4.1 1.4	-9.6 ± 0.3 -9.9 ± 0.3 -9.2 ± 0.4	+24.1±0.4 +23.5±0.4 -0.1±0.7	+2.7±0.6 +2.4±0.4 +2.3±0.9	-12.4 ± 0.4 -12.5 ± 0.4 -12.9 ± 0.5	-1.5 ± 0.5 -1.5 ± 0.9 -0.5 ± 1.0	-9.6 ± 0.3 -9.9 ± 0.3 -9.2 ± 0.4			
C1 S2			+0.1±0.3	$+0.5\pm0.4$	+0.3±0.6	-0.1 ± 0.5	+1.1±0.9	+0.1±0.3			
		Normalized to ¹⁴⁴ Nd/ ¹⁴² Nd; Index Isotope ¹⁴² Nd									
EK1-4-1 SC For $f^* = 20^{\ddagger}$				+28.9±0.4 +38.9±0.4	-2.1 ± 0.6 +27.9 ±0.6	-22.0 ± 0.6 +18.0±0.6	-20.7 ± 0.9 +39.3±0.9	-38.4 ± 1.0 +41.6±1.0			

Deviations relative to the grand mean of terrestrial normals: ${}^{142}Nd/{}^{144}Nd = 1.138266\pm9$; ${}^{145}Nd/{}^{144}Nd = 0.348968\pm6$; ${}^{146}Nd/{}^{144}Nd = 0.724109\pm10$; ${}^{148}Nd/{}^{144}Nd = 0.243079\pm6$; ${}^{150}Nd/{}^{144}Nd = 0.238581$. Standard value of ${}^{143}Nd/{}^{144}Nd = 0.511836$ today for chondritic Sm/Nd. G. Lugmair personal communication. ${}^{\dagger}\epsilon$ 143 variations are also due to decay of ${}^{147}Sm$ ($\tau_{1/2} = 1.06 \times 10^{11}$ years). ${}^{\ddagger}f^{} = ({}^{144}Nd^{*}/{}^{144}Nd) \times 10^{4}$.

masses 134 and 136, while inclusion EK1-4-1 shows large marked negative anomalies at 130, 132, 134, and 136, and a positive anomaly at 137. Using normalization to a standard ¹⁵⁰Nd/¹⁴²Nd value and ¹⁴⁴Nd as the index isotope, we found inclusion C1 to contain normal Nd, while inclusion EK1-4-1 shows marked negative anomalies in Nd at 142, 146, 148, and 150, and a positive anomaly at 145. These observations demonstrate that isotopic anomalies exist in Ba and Nd for several isotopes in some solar-system materials. These isotopic shifts are due to nuclear effects and cannot be attributed to mass fractionation. Normalizations to ¹³⁵Ba/¹³⁸Ba and ¹⁵⁰Nd/¹⁴²Nd were chosen to minimize the propagation of experimental errors and are otherwise arbitrary. The isotopic shifts depend upon the two isotopes (i, j)used for normalization, and the index isotope (m). Generally, the deviation in parts in 10^4 of isotope k is given by ϵ_{ij}^{km} . The fractionation per mass unit α is determined from $\alpha_{ij} = (R_{ij}^M/R_{ij}^0 - 1)/(i - j)$ where M = measured and 0 = standard. For small shifts, the transformation for ϵ to a new index isotope p and a new normalization (u,v) is given by

$$\epsilon_{uv}{}^{kp} = \epsilon_{ij}{}^{km} - \epsilon_{ij}{}^{pm} + (\epsilon_{ij}{}^{vm} - \epsilon_{ij}{}^{um} \frac{(k-p)}{(u-v)}, \quad (2)$$

As the cause of the nonlinear anomalies is obscure, it is more useful to choose a normalization which minimizes the number of isotopes at which anomalies occur. If we normalize to $^{134}Ba/^{138}Ba$ to correct for fractionation, we obtain the results shown in the bottom of Table 1. For sample C1, the only variation outside of error is a deficiency at mass 135. For EK1-4-1, the anomalies are positive and lie at masses 137 and 135. This choice of normalization appears to minimize the number of anomalies for both samples and implies an excess in ¹³⁵Ba and ¹³⁷Ba in one sample and a deficiency in ¹³⁵Ba in the other. If we interpret the shifts in EK1-4-1 as due to the addition of ¹³⁵Ba and ¹³⁷Ba, this requires that the exotic Ba excesses (*) have ${}^{137}Ba^*/{}^{135}Ba^* =$ 1.6. We note that ¹³⁵Ba, ¹³⁷Ba, and ¹³⁸Ba are unshielded and may be produced in an r-process. From the estimates of Seeger, Fowler, and Clayton (1965), (137Ba/

 135 Ba)_r ~ 1.2, which is comparable to the calculated value. However, this value may be subject to substantial changes, using new values for the Ba cross sections (Stroud 1972) together with a detailed calculation of the branching network (Ward, Newman, and Clayton 1976). If the same number of exotic r-process nuclei are added to ¹³⁸Ba as are added at ¹³⁷Ba, this would cause a fractional increase in the ¹³⁸Ba abundance by 1.00018. This would propagate an error in the discrimination calculation of $\epsilon \sim +1.8$ at mass 130, which is well within errors, and a deficiency of $\epsilon \sim -0.9$ at mass 136, which is just within limits of error. No evidence has been found for variation in the abundance of the pprocess isotopes. It does not appear possible to attribute the deficiency at 135 in C1 to the absence of exotic *r*-process material of the same composition inferred for EK1-4-1 owing to the lack of an effect at 137, although this may be obscured by the errors. Alternatively, the ¹³⁵Ba deficiency could result from a deficiency in rprocess relative to s-process contributions, with r-process production ratios shifted from ${}^{137}\text{Ba}*/{}^{135}\text{Ba}* \sim 1.6$. A deficiency in ¹³⁵Ba could occur if there were a late-stage addition of *r*-process nuclei and if Ba were condensed before ¹³⁵Cs ($\tau_{1/2} = 2.3 \times 10^6$ yr) decayed. The defi-ciency in C1 is particularly important, as it implies that the average solar-system value must be made up by an addition of at least 2 parts in 10⁴ of exotic ¹³⁵Ba. The anomalies in EK1-4-1 are not compatible with in situ fission subsequent to crystallization, as the shifts in Ba and Nd are the same for the total sample and the melilite, although Ba/Nd differ by a factor of 3. Further, the fission hypothesis is not in consonance with the large number of other isotopic anomalies for many elements ranging to low Z.

To expose whether the Nd effects are due to addition of *r*-process nuclei, ¹⁴²Nd (shielded) and ¹⁴⁴Nd, which must have a large *s*-process component, are used for normalization. However, the resultant pattern of deviations does not generate positive anomalies for all other isotopes (Table 2). To obtain all positive values, a substantial addition to ¹⁴⁴Nd must be assumed ($f^* =$ 20 parts in 10⁴). To demonstrate general excesses compatible with *r*-process addition, it will be necessary No. 1, 1978

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to obtain data on Sm and other refractory elements in this mass region with two shielded isotopes.

From the data reported here, as well as the wide range of Xe isotopic anomalies, it is reasonable to consider that ¹²⁹I may also have been added in the same process. Assuming that the negative Ba anomaly in C1 was representative of a deficiency in the whole solar nebula prior to the addition of material as found in EK1-4-1, then as $({}^{135}\text{Ba}/{}^{127}\text{I})_{\odot} \sim 0.3$ (Cameron 1973), we obtain ${}^{135}\text{Ba}^{*}/{}^{127}\text{I} \sim 0.5 \times 10^{-4}$. The time scale for production and injection of exotic low-Z elements is $\sim 3 \times 10^6$ years from the interpretation by Lee, Papanastassiou, and Wasserburg (1977, hereafter LPW) regarding the in situ decay of ²⁶Al in the solar nebula. If this time scale also applies to the injected resolution high-Z elements, then any associated ¹²⁹I ($\tau_{1/2} = 1.6 \times 10^7$ years) will not have decayed. Thus, if ¹²⁹I were added along with the exotic nuclei identified here and commensurate in amount with the small Ba deficiency, this could account for the observed ratio of $^{129}I/^{127}I \sim$ 10⁻⁴. Such a model of small last-"minute" injection of *r*-process material is fully compatible with the Xe observations by Reynolds and his co-workers (Reynolds 1960; Podosek 1970). This implies that the observed ¹²⁹Xe effects are not from ¹²⁹I produced by earlier galactic nucleosynthesis and that the ¹²⁹I-²⁴⁴Pu time scale of $\sim 1-2 \times 10^8$ years is an accident due to the injection of newly made exotic ¹²⁹I in a ratio of ¹²⁹I*/ 127 I $\sim 10^{-4}$ (see Schramm and Wasserburg 1970).

Isotopic anomalies have been known for Ne, Kr, and Xe for over 15 years. It now appears that isotopic anomalies are present in solar-system material for O, Ne, Mg, Ca, Kr, Xe, Ba, and Nd. This must be due to incomplete mixing of dust and gas from different sources at the time of formation of early condensates. From the data described here and in the companion *Letter*, it is manifest that the anomalies may extend to much higher atomic number. This may well extend through the transuranics, which could alter the calculation of the time between the termination of nucleosynthesis and the formation of the solar system and could

further contribute anomalies by nuclides with $\tau_{1/2} \ge$ 10⁶ years. The many peculiar isotopic anomalies in \tilde{Xe} are now not isolated but appear to be part of a large family of complex isotopic anomalies extending beyond Z = 60. The complex Xe spectra in residual material in Allende and other carbonaceous chondrites by Anders et al. (1975), Takahashi et al. (1976), and Lewis, Srinivasan, and Anders (1975) have been interpreted as due to superheavy fission and not due to r-process addition. This view has been subject to some discussion because of the enrichment in both neutron-poor and neutron-rich isotopes in the strange Xe which these workers discovered. This matter has been studied extensively by the Chicago group and by Frick (1977) and Frick and Reynolds (1977). In light of the existence of anomalies over a wide spread in Z, it now seems more plausible to consider the Xe anomalies to be the result of a very complex mixture of processes which include *r*-process components and various implantation mechanisms rather than superheavy fission.

The ball game seems to be wide open. The most likely generic model which may explain the observations appears to be an onion-shell supernova explosion followed by injection into the solar nebula, as outlined by Cameron and Truran (1977) to account for the O and Mg data. Such a model can produce elements of high Z. A reliable astrophysical model must await the experimental determination of the yields of exotic nuclei in the region of the rare earths where chemical fractionation will not obscure the relative contributions.

We are grateful to H. Nagasawa for the as yet unique sample of EK1-4-1. M. T. M. thanks D. A. Papanastassiou for his generous advice and introduction to high-precision mass spectrometry. D. J. DePaolo provided timely assistance and gave helpful advice. We thank R. Ward for his interest and advice on p's and q's and s and r. The excitement was communally shared with our calcined colleagues, D. A. Papanastassiou and Typhoon Lee. This work was supported by NSF grant PHY76-83685 and NASA grant NGL 05-002-188.

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