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Isotopic variations in the rock-forming elements in meteorites

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Variations in isotopic abundances of the major rock-forming elements can be used as tracers for chemical processes in the solar nebula, and may also provide links to the presolar cloud from which the solar nebula was derived. Emphasis in this paper is placed on the correlation of isotopic variations between pairs of elements, both for mass-dependent fractionation effects and for nucleosynthetic effects. Variations in oxygen isotope abundances, which are ubiquitous in all Solar System matter, are decoupled from those in other elements, probably because of the effect of a large oxygen reservoir in the nebular gas. Among the metallic rock-forming elements (Mg, Si, Ca, Ti, Cr, Fe, Ni) isotopic variations are small to immeasurable in ordinary chondrites and achondrites. Large variations are observed in refractory inclusions in carbonaceous chondrites in the elements Mg, Si, Ca and Ti. Fractionation effects result from evaporation and condensation at high temperatures. The dominant nucleosynthetic effects are seen as excesses and deficiencies of the neutron-rich isotopes in the region of the iron abundance peak: ⁴⁸Ca, ⁴⁹Ti, ⁵⁰Ti, ⁵⁴Cr, ⁶⁴Ni. These effects result from mixing in different proportions of the products of different regions of a supernova. The rock-forming elements also show isotopic variations due to extinct radioactivities: ²⁶Mg, ⁵³Cr and ⁴¹K. A local source is likely, as is heterogeneous distribution within the solar nebula.

1. INTRODUCTION

Isotopic variations in primitive Solar System materials can occur as a result of physical or chemical fractionation processes, or as a result of nuclear reactions. These variations may be used both for the study of processes in the formation of the Solar System and as probes into stellar nucleosynthesis. It is convenient to group the elements according to their cosmochemical volatility into three categories: (1) the noble gases, (2) the chemically active volatile elements, principally hydrogen, carbon and nitrogen, and (3) the rock-forming elements. Table 1 lists, in order of decreasing Solar System abundance, the twelve most abundant rock-forming elements. Of these, Al, Na, Mn and P have only one stable isotope. Three of these have, however, a cosmochemically important radioactive isotope which has produced isotopic variations in the daughter element: ²⁶Al decays to ²⁶Mg ($t_{1/2} = 0.72$ Ma), ²²Na decays to ²²Ne ($t_{1/2} = 2.6$ a), and ⁵³Mn decays to ⁵³Cr ($t_{1/2} = 3.8$ Ma). The remaining eight elements all have three or more stable isotopes, which makes it possible to discriminate between simple mass-dependent fractionation effects and nuclear or exotic chemical effects.

In this paper we shall review recent developments involving meteoritic abundances of the isotopes of oxygen, magnesium, silicon, calcium, titanium and chromium. Some of the effects

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observed are due to mass-dependent fractionation, and some are due to interaction of isotopically distinct reservoirs, reflecting incomplete homogenization of materials with different nucleosynthetic histories.

Information from isotopic studies can be maximized if several elements are analysed in each sample. Clayton *et al.* (1983*a*, 1985) reported combined oxygen and silicon isotope data on individual chondrules and calcium-aluminium-rich inclusions (CAI) from ordinary and carbonaceous chondrites. Niederer & Papanastassiou (1984) compared isotopic fractionation effects in magnesium and calcium in CAI. These studies showed that processes of evaporation and condensation of silicates led to substantial enrichments and depletions of heavy isotopes of magnesium, silicon, and calcium in CAI. The data rule out an origin of CAI as single-stage condensates from a hot gaseous solar nebula. A single-stage evaporation of an isotopically homogeneous solid precursor to leave a refractory residue is also precluded. Multiple stages of partial evaporation and condensation are required to account for the observed correlations.

Although oxygen and silicon exhibit substantial isotopic variation in chondrules and CAI, the variations are uncorrelated: these two abundant elements had different cosmochemistries in the solar nebula. This lack of correlation is shown in figure 1. It is evident from the solar

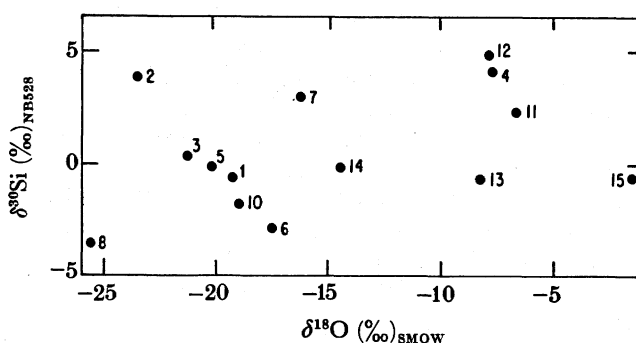


FIGURE 1. Comparison of oxygen and silicon isotopic abundances for several refractory inclusions (CAI) from the Allende carbonaceous chondrite. Each data point represents one inclusion. The absence of a correlation indicates control by different cosmochemical processes: evaporation and condensation for silicon, and exchange with an external gaseous reservoir for oxygen.

TABLE 1. SOLAR SYSTEM ABUNDANCES OF ROCK-FORMING ELEMENTS^a

O	20.1×10^6	S	0.51×10^6	Ni	4.94×10^4
Mg	1.07×10^6	Al	8.49×10^4	Cr	1.34×10^4
Si	1.00×10^6	Ca	6.11×10^4	P	1.04×10^4
Fe	0.90×10^6	Na	5.70×10^4	Mn	0.95×10^4

^a Atomic abundances, relative to Si = 10^6 , from Anders & Ebihara (1982).

abundances in table 1 that much more oxygen is present than the amount required by stoichiometry to convert the other rock-forming elements into rocks. The excess oxygen must have been combined with hydrogen and carbon to form a gaseous reservoir of H₂O and CO. Thus the high-temperature chemistry of oxygen was dominated by isotopic exchange between condensed phases and gas, whereas magnesium, silicon and calcium exhibit the effects of evaporation and condensation. For this reason, oxygen isotopes will be discussed separately from the other rock-forming elements.

2. OXYGEN ISOTOPES

The characteristic systematic patterns of isotopic variation for oxygen are large variations of ^{16}O abundance, with very small variations in $^{17}\text{O}/^{18}\text{O}$. These patterns are interpreted as mixing lines resulting from isotopic exchange between ^{16}O -rich solids (in most cases) and ^{16}O -poor gases (Clayton & Mayeda 1977). The different ^{16}O abundances in the reservoirs may reflect inherited differences of nucleosynthetic components (Clayton *et al.* 1973), or may result from some non-mass-dependent chemical process in the solar nebula (Thiemens & Heidenreich 1983). Whatever the origin, it is clear that oxygen isotopic heterogeneities existed in the early Solar System on scales ranging from micrometers (individual mineral grains) to hundreds of kilometres (meteorite parent bodies) (Clayton *et al.* 1976). Oxygen isotopes can thus serve as natural tracers for a variety of cosmochemical processes. The following is a list of several such applications.

(1) Because the isotopic compositions of individual chondrules and CAI define ^{16}O -mixing lines rather than fractionation lines, they must have been acquired by exchange with an oxygen-bearing nebular gas (CO and/or H_2O). The compositions were therefore established in nebular processes and not parent-body processes (Clayton *et al.* 1985).

(2) At least three isotopically different solid precursor reservoirs are needed to account for the three isotopically distinct populations of chondrules in ordinary chondrites, enstatite chondrites and carbonaceous chondrites (Clayton *et al.* 1986*b*).

(3) Chondrules in unequilibrated ordinary chondrites of iron groups H, L and LL were formed from a single precursor reservoir before the parent bodies were formed (Gooding *et al.* 1983). The isotopic distinction among H, L and LL meteorites may be due to size-sorting of chondrules (Clayton *et al.* 1986*c*).

(4) CAI were formed initially from the most ^{16}O -enriched reservoirs, with mean $\delta^{18}\text{O} = -40\text{‰}$, $\delta^{17}\text{O} = -42\text{‰}$ (Clayton *et al.* 1977).

(5) Petrographically and chemically distinct differentiated meteorites from the same parent body can be recognized by the fact that they fall on a single mass-dependent fractionation line. This criterion has been used to identify lunar meteorites (Mayeda *et al.* 1983), to show that shergottites, nakhlites, and Chassigny have a common parent (Clayton & Mayeda 1983), and to show that eucrites, diogenites, howardites, mesosiderites, pallasites and IIIAB irons may also have a common parent body (Clayton & Mayeda 1983; Clayton *et al.* 1983*b*, 1986*e*). Iron meteorites can be related to stony meteorites by this method (Clayton *et al.* 1983*b*). Certain non-relations have also been revealed by oxygen isotopes: Brachina is not part of the SNC group (Clayton & Mayeda, 1983), and the Eagle Station subgroup of the pallasites was not derived from the same parent as the main-group pallasites (Clayton & Mayeda 1978).

(6) Chemical processes on parent bodies may be studied. Temperatures of thermal metamorphism of ordinary chondrites can be estimated from mineral-pair isotopic fractionation (Onuma *et al.* 1972). Distances of diffusion of oxygen during metamorphism can be measured (Olsen *et al.* 1981). Aqueous alteration on carbonaceous parent bodies can be studied to reveal alteration temperatures (0–100 °C) and water:rock ratios (Clayton & Mayeda 1984).

Three recent studies have used oxygen isotopes to discover the relation between macroscopic meteorites and the population of particles in the Solar System that are smaller than 1 mm. Clayton *et al.* (1986*a*) found that S-type (silicate) deep-sea spherules (Brownlee 1981) in the

size range 200–300 μm probably had pre-terrestrial isotopic compositions like that of the anhydrous component of C2 chondrites. Oxygen isotopic compositions of individual interplanetary dust particles (IDP) can be measured by means of the ion microprobe. McKeegan (1987*a*) has analysed several of the rare refractory IDP and found that their oxygen isotopic compositions lie along the CAI mixing line from C3 chondrites (Clayton *et al.* 1977), with the most ^{16}O -rich sample having $\delta^{18}\text{O} = -41\text{‰}$. Furthermore, $\delta^{18}\text{O}$ values of individual spinel and hibonite grains from the Murchison (C2) chondrite were also found to cluster around the $\delta^{18}\text{O} = -40\text{‰}$ endmember of the CAI mixing line. On the basis of the CAI data alone, it had not been possible to say whether the -40‰ value was uniform throughout the samples (each of which consisted of millions of grains) or whether it was an average of many isotopically 'normal' grains with a few very ^{16}O -rich grains. The data of McKeegan (1987*a*) on individual crystals, both within a meteorite and as IDP, clearly show that the particles with a -40‰ composition were a widespread component of the solar nebula.

McKeegan (1987*b*) has also measured oxygen isotopic compositions of four 'chondritic' IDP and found a wide range in both $\delta^{18}\text{O}$ and $\delta^{17}\text{O}$, with both excesses and deficiencies in ^{16}O relative to Earth. None of the compositions corresponds to values found in known meteorites. Further measurements of IDP are needed in order to recognize relations among them and between them and the meteorites.

3. MAGNESIUM, SILICON, CALCIUM AND TITANIUM ISOTOPES

The elements magnesium, silicon, calcium, and titanium show isotopic variability in meteoritic material due to mass-dependent fractionation and nucleosynthetic anomalies. In addition, magnesium may contain excess ^{26}Mg due to decay of ^{26}Al . In ordinary chondrites and in achondrites, silicon isotopic fractionation up to 1 ‰ per atomic mass unit is observed (Molini-Velsko *et al.* 1986); no isotopic variations have yet been found in these meteorite groups for magnesium, calcium and titanium. It can be concluded that, except for oxygen, most of the rock-forming elements were isotopically homogenized in most meteoritic material. It is remarkable, therefore, that such a profusion of isotopic variability appears in those parts of meteorites, particularly in the carbonaceous chondrites, which have experienced a period of high-temperature processing, as recognized by their chemically refractory nature.

At temperatures in the range 1350–1600 K, solid mineral phases in equilibrium with a gas of solar composition at 10^{-4} atm† include corundum (Al_2O_3), hibonite ($\text{CaAl}_{12}\text{O}_{19}$), melilite ($\text{Ca}_2\text{Al}_2\text{SiO}_7$ – $\text{Ca}_2\text{MgSi}_2\text{O}_7$), spinel (MgAl_2O_4), diopside ($\text{CaMgSi}_2\text{O}_6$), forsterite (Mg_2SiO_4) and perovskite (CaTiO_3) (Grossman & Larimer 1974). Nodules containing some or all of these phases are found in type C2 and C3 carbonaceous chondrites, and are generally labelled as calcium-aluminium-rich inclusions (CAI). The origins of CAI may be as primary condensates from a hot solar nebula (Grossman 1972), as residues from evaporation of chondritic matter (Chou *et al.* 1976), or some combination of evaporation and condensation. Because condensed phases and vapours are involved in these processes, isotopic fractionation of the more volatile rock-forming elements, such as magnesium and silicon, is to be expected, and may be useful in interpreting the thermal histories of CAI. Table 2 is a compilation of data on the isotopic

† 1 atm = 101 325 Pa.

compositions of oxygen, magnesium, silicon, calcium, titanium, chromium and nickel in CAI. Samples are included in table 2 only if isotopic data on more than one element are available.

(a) *Fractionation effects*

If isotopic fractionation is associated with processes of evaporation and condensation, then correlations may be expected for elements of comparable volatility, and different behaviour may be observed for elements of different volatility. Of the elements considered here, magnesium and silicon have similar volatilities in laboratory experiments, and aluminium, calcium and titanium are less volatile (Notsu *et al.* 1978; Hashimoto 1983). Figure 2 shows the correlation in mass-dependent fractionation effects in magnesium and silicon. The good correlation is dominated by data from a few FUN inclusions from Allende and Vigarano. FUN (fractionation plus unknown nuclear effects) inclusions are a subset of CAI having exceptionally large fractionation effects (more than 10 ‰/u for Mg, more than 5 ‰/u for Si) as well as nuclear anomalies in several elements (Lee 1979). The fractionations are typically a factor of two to three greater for magnesium than for silicon. If the fractionations are determined kinetically by an inverse square root of the mass relation, then the species required to give the observed effects are Mg atoms and SiO₂ molecules. Evaporation of silicates produces Mg atoms and both SiO and SiO₂ molecules, the proportions depending on the local state of oxidation (DeMaria *et al.* 1971). Most of the fractionations of magnesium and silicon isotopes are positive (heavy-isotope enrichment), as expected for evaporation residues. Some are negative, indicating condensation from a vapour depleted in heavy isotopes. In general, the 'coarse-grained' CAI are mostly evaporation residues, and 'fine-grained' CAI are mostly condensates.

Figure 3 compares the isotopic fractionations of magnesium and calcium. It is clear that there is no simple correlation between these effects, either in the total population or in any mineralogically selected subgroup. All four quadrants of the figure are occupied, indicating that the fractionations may occur in opposite directions in the same inclusion. These complexities were discussed in detail by Niederer & Papanastassiou (1984), who emphasized the inadequacy of single-stage evaporation or condensation to account for the data. The variability in magnitude and sign of the calcium isotopic fractionation is surprising in light of the very refractory nature of calcium in laboratory vaporization experiments, in which appreciable loss of calcium occurs only after magnesium and silicon are almost completely gone (Hashimoto 1983). It may be that the chemistry of calcium in the solar nebula was more complex, perhaps involving volatile hydroxide species (Hashimoto & Wood 1986).

Figure 4 shows a comparison of magnesium fraction with titanium fractionation. No correlation is evident. All measured titanium fractionations are positive, whereas both positive and negative fractionations occur in magnesium. If heavy-isotope enrichment is a signature of evaporation, it appears that titanium does not re-condense into CAI. It should be noted that all of the samples with large titanium fractionations (greater than 2 ‰/u) are hibonite grains, i.e. much more refractory than most CAI. The absence of a very large heavy-isotope enrichment argues against derivation of the hibonite inclusions simply by evaporation of CAI. Some movement of one or more elements may occur after CAI are formed. Evidence from the systematics of ²⁶Mg excesses (from ²⁶Al decay) correlated with the ²⁷Al/²⁴Mg ratio indicates that both magnesium introduction and exchange may occur (Ireland *et al.* 1986) and this

TABLE 2. ISOTOPIC COMPOSITIONS OF OXYGEN, MAGNESIUM,

(See pages 490–491 for

ii	iii	iv	v	vi	vii	viii	ix	x	xi	xii	xiii	xiv
meteorite	sample	$\delta^{17}\text{O}$	$\delta^{18}\text{O}$	$\delta^{25}\text{Mg}$	$\delta^{26}\text{Mg}$	$^{26}\text{Al}/^{27}\text{Al}$	$^{27}\text{Al}/^{24}\text{Mg}$	$\delta^{30}\text{Si}$	$\delta^{29}\text{Si}$	$\delta^{44/40}\text{Ca}$	$\delta^{42}\text{Ca}$	$\delta^{43}\text{Ca}$
AL	HAL	-12.30	13.70	< 5.00	15.40 (5.50)	5.20×10^{-8} (1.70×10^{-8})	42000	0.00	—	6.70 (0.10)	-1.90 (0.30)	-1.00 (0.50)
AL	TE	-19.34	-6.78	12.90 (0.50)	-0.60 (1.10)	—	2.5	5.96	0.04	-0.60 (0.03)	0.01 (0.05)	0.06 (0.17)
AL	EK-1-4-1	-31.90	-20.80	20.40 (1.40)	-3.60 (0.30)	—	—	6.20	1.30	-1.80 (0.03)	1.70 (0.20)	0.80 (0.30)
AL	Cl	-22.91	-7.11	30.00 (1.10)	-0.60 (0.30)	$< 4.10 \times 10^{-8}$	20	14.26	0.59	0.30 (0.10)	0.20 (0.30)	-0.30 (0.50)
AL	CG-14	-22.09	-10.96	18.30 (0.70)	-1.60 (1.00)	—	2.5	6.80	0.35	—	—	—
VI	1623-5	-23.00	-8.30	—	—	—	—	10.30	—	—	—	—
AL	Egg-1	-16.67	-12.59	0.00	—	—	—	0.51	0.02	0.08 (0.03)	-0.10 (0.10)	0.00 (0.20)
AL	Egg-2	-40.00	-37.88	3.00	—	—	—	-0.13	0.09	-0.18 (0.03)	0.00 (0.20)	0.00 (1.00)
AL	Egg-3	-35.97	-32.32	6.80 (1.30)	78.00 (2.00)	4.90×10^{-8}	148	1.90	0.30	-1.00 (0.02)	-0.40 (0.20)	0.30 (0.60)
AL	Egg-5	—	—	-9.00	—	—	—	-0.30	0.07	0.63 (0.03)	—	—
AL	Egg-6	-41.89	-39.15	—	1.00 (4.00)	—	—	0.03	0.00	-1.60 (0.80)	—	—
AL	F2	-27.54	-23.28	7.90 (0.40)	0.68 (0.36)	—	—	1.85	0.07	—	0.07 (0.09)	0.06 (0.10)
AL	F9	—	—	2.30 (0.30)	1.05 (0.08)	—	—	0.55	—	—	-0.08 (0.06)	0.09 (0.20)
AL	F11	-10.30	-6.60	4.90 (1.30)	1.64 (0.84)	—	—	1.15	-0.08	—	-0.02 (0.06)	0.01 (0.08)
AL	F12	-11.69	-7.83	10.90 (0.30)	1.29 (0.27)	—	—	2.40	0.00	—	-0.04 (0.14)	-0.06 (0.11)
AL	H-18	—	—	2.50 (0.40)	0.64 (0.46)	—	—	—	—	—	-0.11 (0.03)	-0.05 (0.07)
MU	MUR-A1	-49	-47	2.00 (5.00)	36.00 (2.10)	4.00×10^{-8} (4.00×10^{-8})	113.6	4.00 (5.00)	4.00 (11.00)	—	-0.60 (1.80)	-0.70 (2.10)
MU	MUR-H7	-46	-46	-0.80 (5.00)	2.00 (3.20)	$< 4.20 \times 10^{-8}$	77.6	26.00 (22.00)	11.00 (48.00)	—	1.90 (3.30)	1.90 (3.40)
MU	MUR-H8	-43	-39	3.70 (5.00)	1.30 (2.00)	$< 5.70 \times 10^{-8}$	73.5	9.00 (4.00)	-3.00 (9.00)	—	4.40 (3.00)	4.40 (3.40)
MU	MUR-H9	-44.4	-39.9	1.40 (5.00)	0.00 (3.10)	$< 3.40 \times 10^{-8}$	118.1	—	—	—	-0.50 (2.70)	0.20 (3.40)
MU	MUR-20	—	—	-1.10 (1.10)	-2.08 (1.44)	—	171	—	—	—	-1.40 (2.00)	-1.10 (1.00)
MU	MUR-70	—	—	-4.30 (0.20)	-0.35 (0.65)	—	150	—	—	—	-4.40 (3.00)	-3.00 (3.40)
MU	MUR-170	—	—	-2.90 (1.20)	1.97 (1.62)	—	32	—	—	—	2.40 (3.20)	0.80 (3.70)
MY	MY-CH1	—	—	-0.40 (5.00)	0.50 (2.20)	$< 1.00 \times 10^{-8}$	3.5	—	—	—	—	—
MY	MY-H3, 4	-40	-40	-2.00 (5.00)	-0.50 (2.10)	$< 2.40 \times 10^{-8}$	87.2	-12.00 (27.00)	20.00 (70.00)	—	0.20 (2.70)	-1.60 (2.80)
MY	MY-1P	—	—	—	—	—	—	—	—	—	—	—
CB	CB-H2	—	—	2.79 (5.00)	8.80 (1.60)	5.5×10^{-8} (1.30×10^{-8})	19	—	—	—	—	—
CB	CB-H4	—	—	—	—	—	—	—	—	—	—	—
LA	H-H-1	—	—	—	—	—	—	—	—	—	-8.00 (2.80)	-5.20 (3.70)
AL	DA2-12	—	—	2.70 (5.00)	-1.10 (3.50)	$< 3.30 \times 10^{-8}$	102	—	—	—	—	—
MU	TRI31	—	—	10.40 (0.40)	9.74 (0.77)	3.80×10^{-8}	35.4	—	—	—	—	—
MU	TRI52	—	—	—	—	—	—	—	—	—	—	—
MU	TRI54	—	—	-2.45 (0.60)	0.63 (0.76)	—	92	—	—	—	—	—
MU	TRI43	—	—	2.00 (1.81)	-0.36 (0.65)	—	13	—	—	—	—	—
MU	TRI61	—	—	16.90 (0.20)	-2.50 (0.39)	—	31.9	—	—	—	—	—
MU	MH-8	—	—	350.00	—	$< 1.00 \times 10^{-8}$	63	—	—	—	—	—
MU	BB-5	-62.50	-56.60	-7.00 (3.20)	7.00 (1.60)	$< 5.00 \times 10^{-8}$	130	—	—	-3.20 (0.60)	-7.20 (1.30)	-2.60 (1.70)
MU	GR1-C	—	—	-4.40 (1.80)	-0.80 (3.30)	$< 1.20 \times 10^{-8}$	398	—	—	-2.60 (1.90)	—	—
MU	GR1-R	—	—	-6.50 (2.70)	-1.70 (1.80)	$< 1.80 \times 10^{-8}$	137	—	—	-7.30 (1.40)	—	—
MU	SH-7	-26.59	-25.90	1.90 (1.10)	-1.30 (0.90)	$< 3.00 \times 10^{-8}$	222	—	—	6.70 (0.90)	—	—
AL	D3	—	—	1.00	—	—	—	—	—	-2.15 (0.75)	—	—
MU	Pink Angel	—	—	—	—	—	—	—	—	—	—	—
AL	ALK110	—	—	—	—	—	—	—	—	—	0.04 (0.14)	0.13 (0.22)
AL	ALK111	—	—	—	—	—	—	—	—	—	-0.06 (0.07)	-0.04 (0.09)
AL	EK-1-7-1	-38.99	-36.72	—	—	—	—	—	—	—	0.07 (0.10)	0.02 (0.09)
AL	3675A	-19.18	-15.06	9.00	< 1.00	—	—	3.50	-0.03	—	0.09 (0.08)	0.14 (0.20)
AL	EK-1-4-3	—	—	—	—	—	—	—	—	—	-0.11 (0.06)	-0.17 (0.06)
AL	3673	-16.83	-13.17	—	—	—	—	—	—	—	-0.01 (0.06)	-0.08 (0.14)
AL	7R-41	-10.56	-6.39	—	—	—	—	—	—	—	0.02 (0.09)	-0.06 (0.27)
AL	7R-44A1	-19.54	-16.39	—	—	—	—	—	—	—	0.01 (0.16)	-0.01 (0.03)
AL	ALK100	—	—	—	—	—	—	—	—	—	0.02 (0.15)	-0.05 (0.16)
AL	B29	-16.29	-12.59	-9.60	2.60 (0.20)	—	—	-0.64	-0.05	-3.80 (0.03)	-0.40 (0.30)	-0.30 (0.80)
AL	AL15-5	-35.70	-34.80	4.00	—	—	—	0.87	0.16	-0.30 (0.05)	—	—
AL	A2-S1	-22.0	-18.5	1.00	—	—	—	—	—	-0.75 (0.13)	-0.10 (0.10)	0.00 (0.20)
AL	BG10a	—	—	6.30 (0.80)	1.40 (0.20)	—	—	—	—	-0.85 (0.03)	-0.10 (0.20)	0.10 (0.40)
AL	D-2	—	—	—	—	—	—	—	—	-0.10 (0.15)	—	—
AL	D-7	-8.74	-5.34	9.00	—	—	—	—	—	-0.75 (0.03)	0.00 (0.10)	0.00 (0.50)
AL	WA	—	—	5.00	86.00 (0.50)	5.10×10^{-8}	235	—	—	-0.75 (0.03)	-0.20 (0.20)	0.10 (0.30)
AL	A-4	—	—	—	—	—	—	—	—	0.28 (0.05)	0.00 (0.50)	0.00 (0.60)
AL	B30	—	—	-3.00	2.70 (0.20)	—	10.4	—	—	0.93 (0.03)	0.10 (0.10)	-0.30 (0.30)
AL	3598	—	—	0.00	—	—	—	—	—	1.20 (0.05)	0.00 (0.10)	0.00 (0.20)
AL	B32	-10.64	-6.23	1.00	—	—	—	-0.55	0.04	-1.63 (0.03)	-0.10 (0.20)	-0.20 (0.50)
AL	AL-11-PF	—	—	—	—	—	—	—	—	—	—	—
AL	3509	—	—	—	—	—	—	—	—	-0.58 (0.03)	0.00 (0.10)	0.00 (0.10)
LE	1A/LEO1	—	—	6.00	—	—	—	—	—	-0.28 (0.03)	0.00 (0.10)	0.00 (0.20)
AL	3643-1	-15.61	-12.57	—	—	—	—	—	—	—	—	—

IRON, CALCIUM, TITANIUM, CHROMIUM AND NICKEL IN CAI

ferences and footnotes.)

xv	xvi	xvii	xviii	xix	xx	xxi	xxii	xxiii	xxiv	xxv	xxvi
$\delta^{40}\text{Ca}$	$\delta^{48}\text{Ca}$	$\delta^{46/48}\text{Ti}$	$\delta^{47}\text{Ti}$	$\delta^{49}\text{Ti}$	$\delta^{50}\text{Ti}$	$\epsilon^{53}\text{Cr}$	$\epsilon^{54}\text{Cr}$	$\epsilon^{60}\text{Ni}$	$\epsilon^{61}\text{Ni}$	$\epsilon^{64}\text{Ni}$	references
-10.00 (12.00)	-2.20 (0.70)	5.50 (0.90)	-1.80 (3.20)	0.50 (3.60)	14.00 (4.80)	—	—	—	—	—	1, 2, 3, 4, 5, 58
-0.50 (0.80))	0.49 (0.08)	0.70 (0.29)	-0.38 (0.24)	-0.15 (0.26)	0.95 (0.26)	—	—	—	—	—	6, 7, 8, 35, 40
3.30 (1.00)	13.40 (0.20)	0.12 (0.26)	1.32 (0.10)	1.90 (0.19)	3.75 (0.16)	16.20 (0.70)	48.50 (0.50)	—	—	—	2, 8, 18, 19, 28, 52, 53, 58
-10.00 (7.00)	-3.00 (0.30)	0.80 (0.21)	0.57 (0.08)	-0.57 (0.10)	-5.10 (0.11)	2.70 (1.60)	-23.20 (1.80)	—	—	—	2, 8, 11, 19, 28, 40, 52, 53, 58
—	—	—	—	—	—	—	—	—	—	—	6, 40
—	—	—	—	—	—	—	—	—	—	—	59
—	0.60 (0.20)	—	-0.29 (0.09)	-0.46 (0.12)	0.64 (0.09)	—	—	—	—	—	9, 53
—	-0.20 (1.00)	—	—	—	—	—	—	—	—	—	9
—	0.30 (0.20)	0.76 (0.19)	-0.30 (0.08)	-0.45 (0.08)	0.69 (0.10)	-2.00 (1.10)	6.70 (1.30)	—	—	—	6, 8, 9, 28, 39, 52, 56
—	—	—	—	—	—	—	—	—	—	—	61
—	—	0.11 (0.36)	-0.06 (0.20)	0.07 (0.34)	0.69 (0.35)	—	—	—	—	—	8, 9, 39
—	—	0.74 (0.17)	-0.05 (0.07)	-0.34 (0.11)	0.44 (0.12)	-1.40 (0.60)	5.80 (1.20)	—	—	—	8, 17, 28, 33, 39, 52
-3.80 (4.50)	0.85 (0.13)	—	-0.06 (0.05)	+0.02 (0.04)	0.71 (0.08)	—	—	—	—	—	54, 58
1.80 (3.90)	0.19 (0.09)	—	-0.15 (0.08)	-0.03 (0.04)	0.89 (0.08)	—	—	—	—	—	13, 14
-0.50 (3.80)	0.55 (0.11)	—	-0.06 (0.04)	0.01 (0.02)	1.29 (0.08)	—	—	0.20 (0.20)	-0.70 (0.50)	2.00 (0.60)	13, 14, 43
-4.90 (3.20)	0.81 (0.08)	—	-0.15 (0.04)	-0.01 (0.04)	1.25 (0.03)	—	—	—	—	—	13, 14
-1.00 (5.20)	0.65 (0.05)	—	-0.26 (0.06)	-0.06 (0.11)	0.99 (0.15)	—	—	0.00 (0.30)	-0.60 (0.70)	1.40 (0.70)	13, 43
—	4.00 (2.30)	—	-0.50 (1.80)	-0.40 (1.80)	4.00 (2.20)	—	—	—	—	—	4, 15, 23, 26, 30, 47
—	32.70 (3.90)	< 6	0.00 (0.60)	3.00 (1.70)	10.30 (1.80)	—	—	—	—	—	4, 15, 26, 30, 41
—	15.60 (3.30)	< 6	1.10 (2.60)	1.70 (2.20)	1.80 (3.40)	—	—	—	—	—	4, 15, 26, 30, 41
—	20.40 (3.60)	< 6	1.70 (2.00)	-0.20 (2.00)	13.60 (2.90)	—	—	—	—	—	4, 16, 24, 41, 47
—	-45.90 (3.00)	4.20 (0.46)	-1.40 (0.40)	-3.20 (0.70)	-47.20 (0.90)	—	—	—	—	—	15
—	-35.00 (3.50)	8.00 (0.26)	-1.00 (0.30)	-5.00 (0.90)	-29.00 (0.80)	—	—	—	—	—	15, 26
—	-5.80 (3.80)	2.32 (0.26)	-1.30 (0.40)	2.60 (0.60)	-49.60 (0.80)	—	—	—	—	—	15, 26, 46
—	—	< 6	0.30 (1.50)	0.50 (1.40)	3.60 (2.00)	—	—	—	—	—	4, 41, 47
—	51.60 (3.90)	< 6	-1.10 (1.80)	3.10 (1.90)	102.70 (2.90)	—	—	—	—	—	4, 15, 23, 26, 30, 41
—	—	< 6	—	—	—	—	—	—	—	—	47
—	—	< 6	-0.40 (3.00)	-0.20 (2.60)	-0.20 (3.80)	—	—	—	—	—	4, 41
—	—	< 6	-0.20 (1.50)	-0.80 (1.60)	-1.40 (2.60)	—	—	—	—	—	4, 41, 47
—	—	< 6	-0.60 (2.20)	1.60 (2.30)	16.00 (3.20)	—	—	—	—	—	4, 41
—	-33.60 (4.30)	—	-1.20 (2.90)	-9.70 (2.50)	-57.70 (3.40)	—	—	—	—	—	29
—	—	< 6	-0.80 (2.20)	3.90 (2.20)	11.20 (3.00)	—	—	—	—	—	4, 41
—	—	3.93 (0.49)	-1.10 (0.48)	-0.93 (0.76)	0.73 (1.14)	—	—	—	—	—	25, 46
—	—	6.49 (0.24)	1.09 (0.35)	-0.38 (0.53)	-1.14 (0.77)	—	—	—	—	—	25, 46
—	—	2.94 (0.20)	3.09 (0.31)	1.97 (0.36)	3.54 (0.58)	—	—	—	—	—	25, 46, 62
—	—	5.65 (0.39)	1.58 (0.41)	-1.27 (0.57)	-20.73 (0.79)	—	—	—	—	—	25, 46, 62
—	—	6.93 (0.32)	1.04 (0.40)	-3.33 (0.61)	-23.41 (0.84)	—	—	—	—	—	25, 46
—	—	—	1.50 (2.00)	-3.50 (2.00)	1.00 (4.00)	—	—	—	—	—	37
—	-56.10 (3.70)	1.30 (1.00)	-1.30 (1.70)	-10.50 (1.90)	-71.50 (2.30)	—	—	—	—	—	16, 17, 21, 22, 30
—	—	1.10 (1.90)	-1.20 (2.40)	3.10 (2.10)	-5.40 (2.80)	—	—	—	—	—	21, 64
—	—	0.40 (2.90)	-2.30 (1.40)	0.80 (2.00)	-7.10 (4.40)	—	—	—	—	—	21, 64
—	—	4.70 (1.80)	0.10 (1.30)	-0.60 (0.80)	16.90 (1.10)	—	—	—	—	—	21, 63
—	—	0.95 (0.37)	-0.10 (0.11)	-0.17 (0.26)	0.83 (0.20)	—	—	—	—	—	8, 9
—	—	0.37 (0.22)	-0.20 (0.14)	0.06 (0.20)	0.76 (0.13)	-0.20 (0.50)	7.70 (0.70)	—	—	—	8, 28, 52
0.60 (1.30)	0.34 (0.17)	—	-0.31 (0.16)	-0.31 (0.18)	0.32 (0.23)	—	—	1.00 (0.50)	0.00 (0.30)	0.00 (0.50)	7, 12, 50
0.30 (1.20)	0.03 (0.17)	—	-0.11 (0.08)	-0.07 (0.10)	0.75 (0.18)	—	—	1.20 (0.20)	-1.00 (0.50)	2.00 (0.60)	7, 12, 43, 50
0.00 (0.90)	0.29 (0.10)	—	-0.11 (0.13)	0.09 (0.15)	1.02 (0.23)	—	—	—	—	—	7, 10
0.80 (1.20)	0.55 (0.19)	—	0.00 (0.16)	0.41 (0.14)	1.16 (0.23)	—	—	-2.00 (1.50)	0.50 (7.00)	21.00 (10.00)	7, 10, 12
-0.10 (0.90)	0.44 (0.07)	—	—	—	—	—	—	1.00 (0.30)	-0.50 (1.50)	1.00 (2.00)	7, 12
0.50 (1.50)	0.49 (0.13)	—	-0.22 (0.17)	0.00 (0.22)	1.46 (0.35)	—	—	0.80 (0.60)	-0.50 (1.00)	0.00 (1.00)	7, 12, 50
-0.10 (0.70)	-0.06 (0.13)	—	-0.28 (0.15)	-0.13 (0.15)	2.62 (0.23)	—	—	-0.10 (2.50)	-4.00 (16.00)	3.00 (12.00)	7, 10, 12
-0.10 (0.80)	-0.03 (0.28)	—	-0.28 (0.20)	-0.03 (0.16)	0.92 (0.27)	—	—	—	—	—	7, 50
0.40 (0.80)	0.19 (0.15)	—	-0.24 (0.12)	-0.01 (0.15)	0.72 (0.22)	—	—	—	—	—	7, 10
—	-0.20 (0.60)	—	0.00 (0.20)	-0.20 (0.20)	0.52 (0.23)	—	—	—	—	—	9, 40, 60
—	—	—	—	—	—	—	—	—	—	—	9, 40, 60
—	0.30 (0.50)	—	—	—	—	—	—	—	—	—	9, 35
—	0.70 (0.40)	—	—	—	—	—	—	—	—	—	9
—	-0.20 (0.20)	0.18 (0.26)	-0.17 (0.11)	-0.16 (0.13)	1.16 (0.20)	—	—	—	—	—	8, 9, 39, 54
-1.30 (1.30)	-0.10 (0.50)	—	0.05 (0.09)	-0.34 (0.29)	0.24 (0.19)	—	—	—	—	—	2, 9, 31, 54
—	-0.20 (0.60)	—	—	—	—	—	—	—	—	—	9
—	-0.10 (0.20)	0.79 (0.20)	0.00 (0.09)	-0.13 (0.16)	0.74 (0.16)	—	—	—	—	—	9, 34, 54, 55
-1.60 (2.00)	-0.20 (0.20)	—	—	—	—	—	—	—	—	—	9
7.20 (1.30)	0.20 (0.30)	—	0.02 (0.12)	-0.04 (0.12)	0.86 (0.21)	-0.40 (0.70)	6.50 (0.80)	—	—	—	9, 28, 52, 60
—	—	—	-0.09 (0.14)	-0.06 (0.13)	0.95 (0.22)	—	—	—	—	—	10
—	-0.80 (0.20)	—	—	—	—	—	—	—	—	—	9
5.00 (3.00)	0.30 (0.20)	0.25 (0.25)	-0.03 (0.16)	0.04 (0.23)	0.89 (0.21)	—	—	—	—	—	8, 9
—	—	—	-0.03 (0.14)	-0.03 (0.17)	0.70 (0.26)	—	—	—	—	—	10

TABLE 2

ii	iii	iv	v	vi	vii	viii	ix	x	xi	xii	xiii	xiv
meteorite	sample	$\delta^{17}\text{O}$	$\delta^{18}\text{O}$	$\Delta^{25}\text{Mg}$	$\delta^{26}\text{Mg}$	$^{26}\text{Al}/^{27}\text{Al}$	$^{27}\text{Al}/^{24}\text{Mg}$	$\Delta^{30}\text{Si}$	$\delta^{29}\text{Si}$	$\Delta^{44/40}\text{Ca}$	$\delta^{42}\text{Ca}$	$\delta^{43}\text{Ca}$
MU	DJ-1	—	—	—2.20 (2.60)	1.90 (3.30)	$< 1.50 \times 10^{-4}$	—	—	—	0.90 (3.30)	—	—
MU	MUR-HI	-47.5	-48.8	—	—	—	—	—	—	—	3.20 (3.20)	3.20 (3.30)
AL	AL3S4	-39.8	-38.2	—	140.00	4.70×10^{-5} (3.00×10^{-5})	400	1.00	0.00	—	—	—
AL	F7	-19.82	-16.12	4.90 (0.60)	0.84 (0.23)	—	—	1.49	0.05	—	0.04 (0.19)	-0.01 (0.15)
AL	F6	-21.15	-17.44	1.70 (0.60)	0.45 (0.09)	—	—	-1.42	0.03	—	0.06 (0.04)	0.01 (0.09)
AL	F3	-24.59	-21.17	5.20 (0.06)	0.39 (0.27)	—	—	0.17	0.00	—	0.03 (0.06)	-0.11 (0.18)
AL	4691-A	—	—	—	—	—	—	—	—	—	—	—
MU	TR155	—	—	2.40 (0.50)	-7.21 (0.88)	—	193	—	—	—	—	—
AL	3529-40	—	—	—	—	—	—	—	—	—	—	—
AL	A8	—	—	—	—	—	—	—	—	—	—	—
LE	2628	-18.21	-13.37	—	—	—	—	2.40	0.01	—	—	—
AL	D8	-17.18	-13.53	—	—	—	—	1.07	0.00	—	—	—
AL	Egg-4	-13.72	-10.39	—	—	—	—	1.83	-0.01	—	—	—
AL	5241	—	—	—	0.00 (2.00)	—	32	—	—	—	—	—
AL	BG2-6	—	—	0.50	13.10 (0.20)	5.80×10^{-5}	32	—	—	—	—	—
AL	AA1	—	—	—	4.90 (0.30)	5.30×10^{-5}	13	—	—	—	—	—
AL	3529Z	—	—	—	55.00 (6.00)	4.03×10^{-5} (4.70×10^{-6})	190	—	—	—	—	—
AL	ALTL-1	—	—	—	—	—	—	—	—	—	—	—
AL	ALTL-19	—	—	—	—	—	—	—	—	—	—	—
MU	DJ-6	—	—	—	0.00 (2.00)	2.30×10^{-4}	123	—	—	—	—	—
MU	DJ-5	—	—	—	—	—	—	—	—	—	—	—
AL	BG82D1	—	—	-12.20 (5.30)	30.30 (9.80)	—	14	—	—	—	—	—
DH	DH-H1	—	—	19.00 (5.00)	920	8.40×10^{-6} (5.0×10^{-7})	15600	—	—	12.50 (1.20)	—	—
AL	AL-5	—	—	—	—	—	—	—	—	—	—	—
EF	E2	—	—	—	—	—	—	—	—	—	—	—
AL	F1	-22.56	-19.15	0.90 (0.60)	1.76 (0.77)	—	—	-0.29	0.04	—	—	—
AL	F5	-23.62	-20.09	1.40 (0.30)	0.38 (0.54)	—	—	-0.05	0.03	—	-0.06 (0.05)	0.09 (0.20)
AL	F8	-29.26	-26.04	2.30 (0.60)	0.56 (0.16)	—	—	-1.77	0.05	—	-0.03 (0.07)	0.05 (0.09)
AL	F15	-4.86	-1.48	1.50 (0.60)	—	—	—	-0.32	0.00	—	0.01 (0.18)	0.03 (0.19)
AL	BG82DH8	—	—	31.10 (1.00)	-1.60 (0.20)	—	—	—	—	0.40 (0.10)	-0.13 (0.11)	0.06 (0.21)
AL	B7F6	—	—	12.10 (0.80)	-0.80 (0.10)	—	—	—	—	—	-0.12 (0.13)	-0.06 (0.23)
AL	BR-1	—	—	—	—	—	—	—	—	—	—	—
AL	BR-9	—	—	—	—	—	—	—	—	—	—	—
AL	G1	—	—	—	—	—	—	—	—	—	—	—
AL	G3	—	—	—	—	—	—	—	—	—	—	—
AL	G2	—	—	—	—	—	—	—	—	—	—	—
AL	G6	—	—	—	—	—	—	—	—	—	—	—
AL	G4	—	—	—	—	—	—	—	—	—	—	—
LE	LEO-6	—	—	—	—	—	—	—	—	—	—	—
MU	7-51-1	—	—	-5.00 (0.50)	4.00 (0.69)	—	127	—	—	—	—	—
MU	7-48-2	—	—	-2.60 (0.40)	2.56 (0.59)	—	108	—	—	—	—	—
MU	7-29-1	—	—	-1.90 (0.70)	-1.38 (0.95)	—	149	—	—	—	—	—
MU	7-19-1	—	—	0.20 (0.80)	6.04 (0.75)	—	15	—	—	—	—	—
MU	7-143-1	—	—	-3.20 (0.30)	2.73 (0.46)	—	5	—	—	—	—	—
MU	7-412-1	—	—	-6.10 (1.30)	16.05 (0.83)	—	40	—	—	—	—	—
MU	7-664-1	—	—	0.30 (0.30)	3.47 (0.55)	—	6	—	—	—	—	—
MU	7-658-1	—	—	-2.20 (0.30)	4.58 (0.97)	—	66	—	—	—	—	—
MU	7-644-1	—	—	-0.30 (0.50)	11.07 (0.98)	—	20	—	—	—	—	—
MU	7-753-1	—	—	-2.60 (0.50)	-4.74 (1.14)	—	31	—	—	—	—	—
MU	7-789-1	—	—	0.70 (0.70)	12.72 (1.08)	—	37	—	—	—	—	—
MU	7-981-1	—	—	-0.20 (0.40)	0.81 (0.91)	—	139	—	—	—	—	—
MU	7-980-1	—	—	-0.90 (0.30)	1.87 (0.74)	—	152	—	—	—	—	—
MU	7-953-1	—	—	-1.10 (0.40)	5.20 (0.62)	—	14	—	—	—	—	—
AL	3675B	—	—	—	—	—	—	1.80	-0.08	—	0.04 (0.06)	-0.06 (0.07)

Footnotes by column number:

- ii) meteorite abbreviations: AL, Allende; CB, Cold Bokkeveld; DH, Dhajala; EF, Efremovka; LA, Lancé; LE, Leoville; MU, Murchison; MY, Murray; VI, Vigarano;
- iv), (v) $\delta^{17}\text{O}$, $\delta^{18}\text{O}$ as permil (‰) deviations from SMOW standard;
- vi) $\Delta^{25}\text{Mg}$: isotopic fractionation, in per mille per atomic mass unit, with respect to standard $^{25}\text{Mg}/^{24}\text{Mg} = 0.12663$;
- vii) maximum $\delta^{26}\text{Mg}$, excess ^{26}Mg , in ‰, with respect to standard $^{26}\text{Mg}/^{24}\text{Mg} = 0.13938$, after correction for fractionation based on $\Delta^{25}\text{Mg}$;
- viii) initial $^{26}\text{Al}/^{27}\text{Al}$, based on the slope of Al-Mg isochron;
- ix) maximum measured $^{27}\text{Al}/^{24}\text{Mg}$ ratio;
- x) $\Delta^{30}\text{Si}$: isotopic fractionation, in per mille per atomic mass unit, relative to NBS 28 quartz, based on $^{30}\text{Si}/^{28}\text{Si}$ measurements;
- xi) $\delta^{29}\text{Si}$, excess ^{29}Si , relative to NBS 28 quartz, after correction for fractionation based on $\Delta^{30}\text{Si}$;
- xii) $\Delta^{44/40}\text{Ca}$: isotopic fractionation, in per mille per atomic mass unit, with respect to terrestrial standard $^{44}\text{Ca}/^{40}\text{Ca} = 0.021208$;
- xiii) $\delta^{42}\text{Ca}$, excess ^{42}Ca , with respect to terrestrial standard, after correction for fractionation based on $\Delta^{44/40}\text{Ca}$;
- xiv)-(xvi) $\delta^{43}\text{Ca}$, $\delta^{46}\text{Ca}$, $\delta^{48}\text{Ca}$ defined by analogy to $\delta^{42}\text{Ca}$ (column xiii);
- xvii) $\Delta^{46/48}\text{Ti}$: isotopic fraction, in per mille per atomic mass unit, relative to terrestrial standard $^{46}\text{Ti}/^{48}\text{Ti} = 0.108548$;
- xviii)-(xx) $\delta^{47}\text{Ti}$, $\delta^{49}\text{Ti}$, $\delta^{50}\text{Ti}$ defined by analogy to $\delta^{42}\text{Ca}$ (column xiii);
- xxi) $\epsilon^{53}\text{Cr}$: excess ^{53}Cr , in parts in 10^4 with respect to terrestrial standard, after correction for fractionation based on $^{50}\text{Cr}/^{52}\text{Cr}$;
- xxii) $\epsilon^{54}\text{Cr}$ defined by analogy to $\epsilon^{53}\text{Cr}$ (column xxi);
- xxiii)-(xv) $\epsilon^{60}\text{Ni}$, $\epsilon^{61}\text{Ni}$, $\epsilon^{64}\text{Ni}$: excesses in parts in 10^4 with respect to terrestrial standard, after correction for fractionation based on $^{62}\text{Ni}/^{58}\text{Ni}$.

ont.)

xv	xvi	xvii	xviii	xix	xx	xxi	xxii	xxiii	xxiv	xxv	xxvi
$\delta^{46}\text{Ca}$	$\delta^{48}\text{Ca}$	$\delta^{48/46}\text{Ti}$	$\delta^{47}\text{Ti}$	$\delta^{49}\text{Ti}$	$\delta^{50}\text{Ti}$	$\epsilon^{53}\text{Cr}$	$\epsilon^{54}\text{Cr}$	$\epsilon^{60}\text{Ni}$	$\epsilon^{61}\text{Ni}$	$\epsilon^{64}\text{Ni}$	references
—	17.30 (4.30)	4.00 (1.60)	-0.20 (1.10)	-1.10 (2.80)	-20.60 (4.10)	—	—	—	—	—	17, 21
—	—	—	0.60 (1.50)	5.20 (1.40)	14.00 (2.1)	—	—	—	—	—	16
—	—	—	-0.18 (0.43)	-0.42 (0.42)	2.28 (0.84)	—	—	—	—	—	4, 20, 60
0.30 (3.60)	0.41 (0.09)	—	-0.03 (0.09)	-0.01 (0.02)	0.36 (0.09)	—	—	—	—	—	57, 58
-4.40 (2.10)	0.39 (0.18)	—	-0.06 (0.18)	0.12 (0.09)	1.30 (0.18)	—	—	—	—	—	57, 58
-8.40 (13.5)	0.55 (0.14)	—	-0.04 (0.09)	0.75 (0.10)	1.18 (0.15)	-0.55 (0.18)	0.46 (0.30)	—	—	—	57, 58
—	—	—	-0.24 (0.16)	0.04 (0.20)	0.86 (0.31)	—	—	—	—	—	50
—	—	2.95 (0.70)	0.89 (0.65)	6.21 (1.25)	16.50 (1.41)	—	—	—	—	—	15, 25
—	—	—	0.14 (0.35)	-0.18 (0.40)	1.45 (0.33)	—	—	—	—	—	54
—	—	—	—	—	—	0.40 (0.50)	1.20 (1.00)	—	—	—	28, 52
—	—	—	—	—	—	—	—	—	—	—	24
—	—	—	0.01 (0.05)	0.04 (0.07)	1.13 (0.08)	—	—	—	—	—	55, 58
—	—	-0.01 (0.15)	-0.08 (0.08)	0.01 (0.09)	0.92 (0.09)	—	—	—	—	—	54, 55
—	—	—	—	—	—	—	—	—	—	—	33
—	—	—	—	—	—	—	—	—	—	—	34
—	—	—	—	—	—	—	—	—	—	—	34
—	—	—	0.00 (0.13)	0.03 (0.10)	0.86 (0.26)	-4.00 (3.00)	4.00 (4.00)	—	—	—	32, 36, 50
—	—	—	—	—	—	0.00 (2.00)	10.00 (4.00)	—	—	—	36
—	—	—	—	—	—	3.00 (2.00)	13.00 (7.00)	—	—	—	36
—	—	—	—	—	-8.00 (2.00)	—	—	—	—	—	37
—	—	—	—	—	-16.00 (3.00)	—	—	—	—	—	37
—	—	10.00	—	—	—	—	—	—	—	—	38
—	—	—	—	—	—	—	—	—	—	—	3, 41, 64
—	—	—	-0.04 (0.05)	-0.02 (0.07)	0.90 (0.09)	—	—	—	—	—	42
—	—	—	—	—	—	-0.45 (0.13)	6.20 (0.30)	-0.10 (0.20)	-0.90 (0.60)	0.50 (0.70)	14, 43, 57
8.90 (8.90)	0.44 (0.17)	—	-0.13 (0.10)	0.08 (0.08)	1.48 (0.20)	—	—	-0.40 (0.20)	-1.00 (0.50)	1.20 (0.60)	14, 43, 57
1.50 (3.00)	0.55 (0.29)	—	-0.12 (0.14)	-0.09 (0.09)	1.22 (0.18)	-0.62 (0.18)	7.60 (0.30)	0.40 (0.30)	0.50 (0.80)	-0.60 (1.30)	14, 43, 57
-2.70 (3.00)	0.34 (0.07)	—	—	—	—	—	—	0.50 (0.30)	0.90 (0.80)	-0.20 (1.00)	14, 43, 58
-1.00 (5.00)	-3.10 (0.21)	—	0.64 (0.08)	-0.45 (0.08)	-5.00 (0.12)	—	—	—	—	—	44, 45
1.70 (6.60)	-3.19 (0.39)	—	—	—	—	—	—	—	—	—	44, 45
—	—	—	—	—	—	-0.86 (0.73)	4.86 (1.44)	1.70	-2.80 (13.00)	-10.40 (22.0)	48, 49, 51
—	—	—	—	—	1.70 (0.25)	-1.06 (0.36)	6.82 (1.10)	0.50	11.30 (7.40)	11.40 (11.00)	49, 51
—	—	—	—	—	1.08 (0.21)	-1.08 (0.33)	7.31 (1.10)	2.10	-0.80 (8.10)	3.30 (12.00)	49, 51
—	—	—	—	—	—	-1.00 (0.62)	0.50 (1.40)	—	—	—	48
—	—	—	—	—	—	-1.23 (0.50)	5.89 (1.26)	—	—	—	49
—	—	—	—	—	—	1.70 (0.19)	3.45 (0.46)	—	—	—	49
—	—	—	—	—	—	-0.47 (0.45)	6.06 (1.00)	—	—	—	49
—	—	0.10 (0.17)	0.03 (0.07)	0.03 (0.10)	0.60 (0.10)	—	—	—	—	—	55
—	—	6.22 (0.49)	-1.37 (0.41)	-3.52 (0.85)	-25.42 (1.02)	—	—	—	—	—	46, 62
—	—	6.80 (0.44)	-1.45 (0.36)	-3.36 (0.97)	-25.43 (1.51)	—	—	—	—	—	46, 62
—	—	6.13 (0.40)	0.76 (0.53)	-0.14 (0.94)	-11.06 (1.12)	—	—	—	—	—	46, 62
—	—	0.25 (0.56)	-0.29 (0.68)	1.04 (1.01)	2.86 (1.05)	—	—	—	—	—	46, 62
—	—	3.57 (0.32)	-0.48 (0.45)	0.11 (0.82)	3.15 (0.76)	—	—	—	—	—	46, 62
—	—	6.80 (0.31)	-1.10 (0.43)	-2.02 (0.70)	1.55 (0.88)	—	—	—	—	—	46, 62
—	—	2.60 (0.42)	-1.40 (0.47)	-2.87 (0.85)	-1.41 (1.11)	—	—	—	—	—	46, 62
—	—	3.79 (0.29)	-2.10 (0.52)	-5.36 (0.64)	2.04 (0.79)	—	—	—	—	—	46, 62
—	—	4.17 (0.31)	-0.04 (0.55)	0.45 (0.86)	1.09 (1.01)	—	—	—	—	—	46, 62
—	—	5.18 (0.41)	-0.80 (0.50)	2.02 (0.85)	11.56 (1.22)	—	—	—	—	—	46, 62
—	—	4.45 (0.23)	-0.93 (0.34)	-1.85 (0.56)	-1.15 (0.87)	—	—	—	—	—	46, 62
—	—	6.47 (0.19)	0.06 (0.43)	-3.31 (0.68)	-16.09 (0.71)	—	—	—	—	—	46, 62
—	—	5.88 (0.40)	-0.58 (0.55)	-3.91 (1.11)	-16.37 (1.24)	—	—	—	—	—	46, 62
—	—	5.07 (0.26)	-1.21 (0.54)	-1.90 (0.87)	-0.59 (1.18)	—	—	—	—	—	46, 72
-0.03 (0.90)	0.44 (0.12)	—	—	—	—	—	—	—	—	—	7, 68

references:

(1) Lee *et al.* (1980); (2) Lee *et al.* (1979); (3) Hinton & Bischoff (1984); (4) Fahey *et al.* (1987a); (5) R. W. Hinton (personal observation); (6) Hinton *et al.* (1984); (7) Jungck *et al.* (1984); (8) Niederer *et al.* (1985a); (9) Niederer & Papanastassiou (1984); (10) Niemeyer & Lugmair (1984); Esat *et al.* (1978); (12) Shimamura & Lugmair (1983); (13) Prombo & Lugmair (1986); (14) Clayton *et al.* (1985); (15) Zinner *et al.* (1986b); Zinner *et al.* (1988); (17) Hinton *et al.* (1985); (18) Clayton & Mayeda (1977); (19) Wasserburg *et al.* (1977); (20) Steele *et al.* (1978); (21) Hinton *et al.* (1987); (22) Bar-Matthews *et al.* (1982); (23) Fahey *et al.* (1985a); (24) Clayton *et al.* (1986d); (25) Ireland *et al.* (1985); (26) Zinner (1986a); (27) Lee & Zeff (1981); (28) Papanastassiou & Wasserburg (1986); (29) Fahey *et al.* (1986b); (30) Fahey *et al.* (1987b); (31) Lee (1977); (32) Huneke *et al.* (1982); (33) Hutcheon *et al.* (1985); (34) Lee *et al.* (1976); (35) Papanastassiou & Wasserburg (1983); (36) Lee *et al.* (1986); (37) Hutcheon *et al.* (1983); (38) Brigham *et al.* (1986); (39) Niederer *et al.* (1985a,b); (40) Molini-Velsko *et al.* (1983); (41) E. er (personal communication); (42) Niemeyer (1986a); (43) Birck & Lugmair (1987); (44) Brigham *et al.* (1987); (45) Papanastassiou & Brigham (1987); (46) Ireland (1987); (47) Fahey *et al.* (1986a); (48) Birck & Allègre (1984); (49) Birck & Allègre (1985); (50) Niemeyer & Lugmair (1981); Morand & Allègre (1983); (52) Papanastassiou (1986); (53) Niederer *et al.* (1980); (54) Niederer *et al.* (1981); (55) Kastenmeyer *et al.* (1985); R. N. Clayton (1987); (57) Prombo *et al.* (1987); (58) Molini-Velsko (1983); (59) Clayton *et al.* (1987); (60) Clayton *et al.* (1977); (61) Wasserburg (1985); (62) T. R. Ireland (personal communication); (63) Hashimoto *et al.* (1986); (64) Hinton *et al.* (1988).

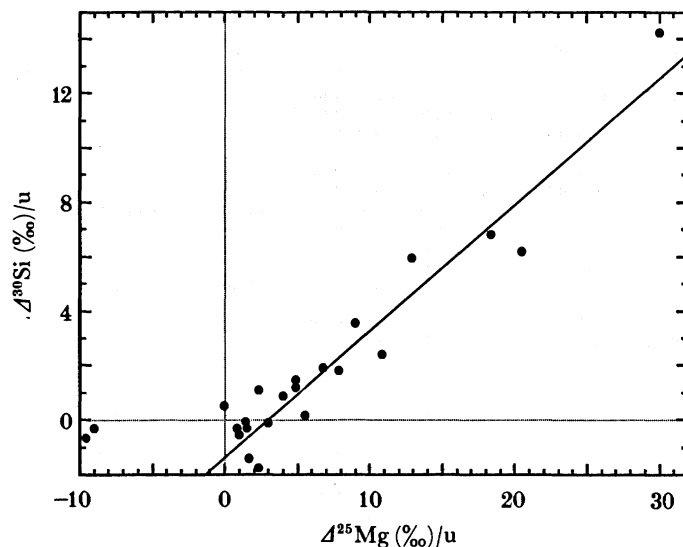


FIGURE 2. Comparison of the mass-dependent isotope fractionations in magnesium and silicon in CAI, expressed as per mille per atomic mass unit. The good correlation results from the similar volatility of the two elements under nebular conditions. The data are dominated by positive Δ values characteristic of evaporation residues.

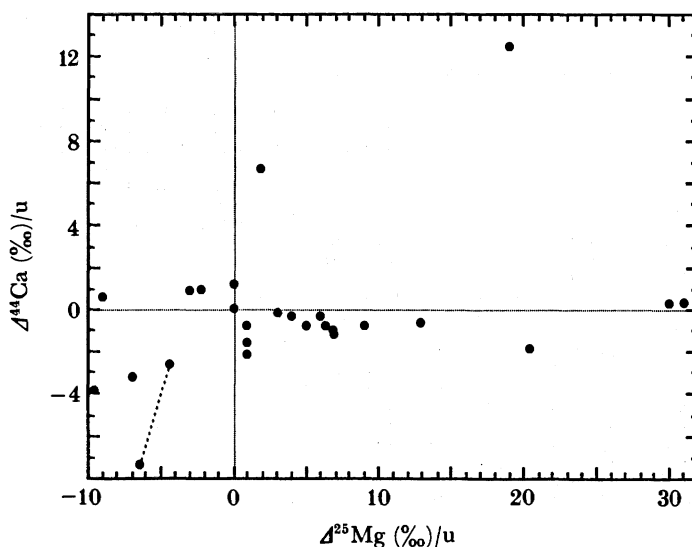


FIGURE 3. Comparison of the mass-dependent isotope fractionations in magnesium and calcium in CAI. The lack of correlation results from the difference in volatility between the two elements, and suggests more complicated chemical behaviour for calcium. The points connected by a dashed line represent rim and core of inclusion GR-1, showing the rim to be depleted in heavy isotopes of calcium and magnesium relative to the core.

exchange may occur before ^{26}Al has decayed (Fahey *et al.* 1985*b*). Perfect correlations between individual elements are therefore not to be expected.

(b) Nucleosynthetic effects

The most prominent nucleosynthetic isotope effects in the rock-forming elements (aside from the ^{16}O excesses) are the excesses and deficiencies of the neutron-rich isotopes in elements of the iron peak. These are now well documented for ^{48}Ca , ^{49}Ti , ^{50}Ti , ^{54}Cr and ^{64}Ni . All of these

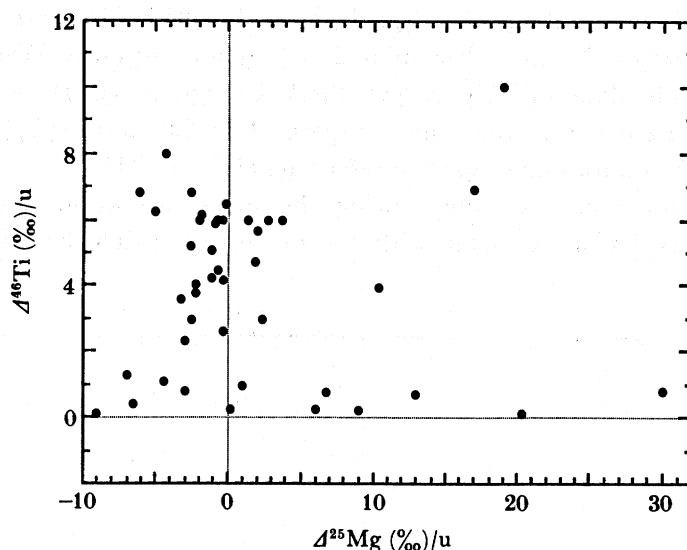


FIGURE 4. Comparison of the mass-dependent isotope fractionations in magnesium and titanium. All titanium fractionations are positive. Samples with $\Delta^{46}\text{Ti} > 2\text{‰}$ are all hibonite, and may contain secondary magnesium.

nuclides can be produced in a neutron-rich e-process (e represents equilibrium) (Cameron 1979; Hartmann *et al.* 1985), believed to occur in supernova explosions. The calculations of Hartmann *et al.* (1985) predict isotopic anomalies of comparable magnitude in ^{48}Ca , ^{50}Ti and ^{54}Cr (normalized to solar abundances), of greater magnitude in ^{66}Zn and ^{84}Kr , and of lesser magnitude in ^{68}Zn and ^{82}Se . Of course, the magnitude of the anomaly ultimately observed in a meteorite sample depends on the chemical fractionations of the material en route from the supernova to the solar nebula. Hence it is not surprising that anomalies in volatile elements such as zinc, selenium and krypton have not been found in CAI. Those anomalies that are observed show at least qualitative correlation from one element to another: see figure 5 for a comparison

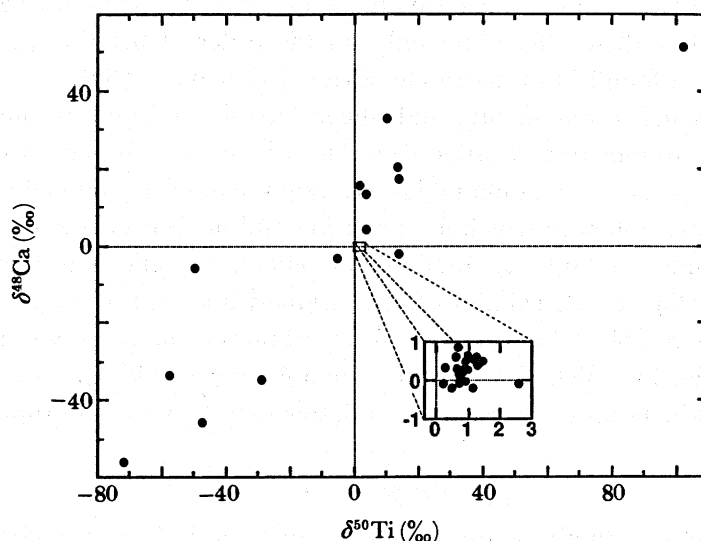


FIGURE 5. Comparison of the nucleosynthetic anomalies in ^{48}Ca and ^{50}Ti . The correlation results from production of these nuclides together in a neutron-rich equilibrium process. Chemical effects introduce scatter in the correlation.

of ^{48}Ca and ^{50}Ti anomalies (Prombo *et al.* 1987; Fahey *et al.* 1986*b*; Zinner *et al.* 1986*b*), and figure 6 for a comparison of ^{50}Ti and ^{54}Cr anomalies (Papanastassiou & Wasserburg 1986; Papanastassiou 1986). The data for ^{64}Ni are few (Birck & Lugmair 1987), but also correlate in sign with ^{50}Ti measurements on the same samples. D. D. Clayton (1987) has presented plausible mechanisms for establishing separate reservoirs of ^{48}Ca , ^{50}Ti , ^{54}Cr on the one hand and the remaining isotopes on the other during the supernova event and subsequently reassembling these reservoirs in CAI, some with excesses of the n-rich material, others with deficits.

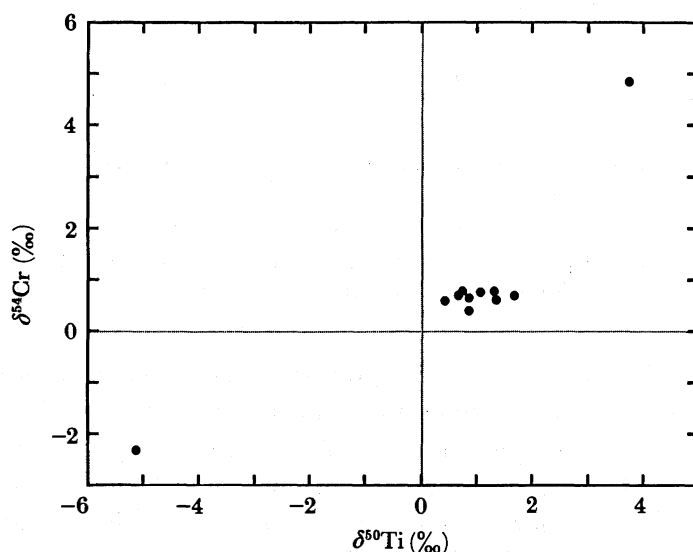


FIGURE 6. Comparison of the nucleosynthetic anomalies in ^{54}Cr and ^{50}Ti . Both nuclides are produced in the neutron-rich equilibrium process.

Anomalies in ^{50}Ti are found not only in CAI, also in whole-rock samples of carbonaceous chondrites (Niemeyer 1985) and in some individual chondrules from carbonaceous chondrites (Niemeyer 1986*a,b*), but these effects are only on the order of 0.1–0.3 ‰. No confirmed anomalies have yet been found in ordinary chondrites (Niemeyer 1987).

Small nuclear anomalies in magnesium and silicon have been found in some Allende CAI, particularly the FUN inclusions (see negative values for $\delta^{26}\text{Mg}$ and non-zero values of $\delta^{29}\text{Si}$ in table 2). It is not possible to make an unambiguous assignment of the anomaly to a particular isotope in a three-isotope system in which fractionation and nuclear effects are of comparable magnitude. For example, in table 2, fractionation effects for silicon are calculated from variations in the $^{30}\text{Si}/^{28}\text{Si}$ ratio. Deviations from a slope- $\frac{1}{2}$ line on a three-isotope graph are given as excesses of ^{29}Si in table 2. However, the fractionations could equally well be calculated from the $^{29}\text{Si}/^{28}\text{Si}$ ratio, in which case the nuclear anomaly would appear as ^{30}Si deficits. Systematics of the FUN inclusions suggest that the latter may be more appropriate.

(c) Radiogenic ^{26}Mg

The existence of nucleosynthetic isotope anomalies in several of the rock-forming elements implies incomplete homogenization, on some scale, of the products of nucleosynthesis from diverse sources. No specific inferences can be drawn about the time of the nucleosynthetic

events relative to the beginning of formation of the Solar System. Information on timing comes from the existence of decay products of short-lived radioactivities, particularly ^{26}Mg from ^{26}Al , ^{107}Ag from ^{107}Pd , ^{129}Xe from ^{129}I , and fission products from ^{244}Pu . Also potentially important are ^{41}K from ^{41}Ca , ^{53}Cr from ^{53}Mn , and ^{60}Ni from ^{60}Fe . Of these, the system on which most attention has focused is the ^{26}Al – ^{26}Mg system, because of the clear signal of excess ^{26}Mg in primitive meteorites, and because of the short half-life of ^{26}Al (0.72 Ma). Of fundamental importance are the questions of whether ^{26}Al was live in the solar nebula, and whether it was uniformly distributed (i.e. constant $^{26}\text{Al}/^{27}\text{Al}$, except for radioactive decay). The former question is important to establish the time between nucleosynthesis and the formation of solid objects in the Solar System; the latter question concerns the importance of ^{26}Al as a heat source in newly formed planetesimals (Urey 1955; Lee *et al.* 1976). To prove that ^{26}Al was once present in a rock sample (a CAI, for example), it is necessary to demonstrate a linear relation between $^{26}\text{Mg}^*/^{24}\text{Mg}$ and $^{27}\text{Al}/^{24}\text{Mg}$ among different phases of the rock. This has been done for many CAI in carbonaceous chondrites (Hutcheon *et al.* 1986) and for one hibonite grain in an ordinary chondrite (Hinton & Bischoff 1984). However, the initial $^{26}\text{Al}/^{27}\text{Al}$ ratios inferred from the slopes of the isochrons vary from a commonly observed value of 5×10^{-5} to values less than 10^{-7} . Several examples are known of hibonite samples with large Al/Mg ratios and no detectable radiogenic ^{26}Mg (e.g. BB5, MUR H-7, MUR H-8, MUR H-9, MY H-3, and MY H-4 in table 2). If these differences result from crystallization after different decay intervals from an initially uniform source, then time intervals of millions of years are required between different CAI in a single meteorite (and sometimes within a single CAI). Several authors (e.g. Hutcheon 1982; Fahey *et al.* 1987*a*) have concluded that such time differences are unreasonably large and that initial isotopic heterogeneities existed, but postulated no mechanism for their origin.

Potentially of great importance for understanding the significance of ^{26}Al in meteorites is the observation of live ^{26}Al in the interstellar medium (ISM) today, based on measurements of the γ -ray emitted in its decay (Mahoney *et al.* 1984). From the γ -ray intensity, Clayton & Leising (1987) estimate the $^{26}\text{Al}/^{27}\text{Al}$ ratio in the ISM to be about 1×10^{-5} , with about a factor of two uncertainty due to lack of knowledge of the spatial distribution of ^{26}Al in the Galaxy. They conclude that this ratio is too high to be maintained by ^{26}Al produced in supernovae, and thus rule them out as a principal source of ^{26}Al , and propose novae as the source. They also conclude that the ratio is too low to account for the value of 5×10^{-5} commonly found in meteoritic CAI. They further believe that the radiogenic ^{26}Mg in meteorites was not formed *in situ* from live ^{26}Al , but is a 'fossil' formed in grains in interstellar space, which, by a series of selective chemical reactions, became mixed into CAI in such a way as to mimic isochrons.

Cameron (1984) accepts the evidence for *in situ* decay of ^{26}Al , ^{107}Pd , and ^{129}I in the Solar System, as well as the arguments against a supernova and the inadequacy of the ISM levels of ^{26}Al , and seeks an alternative local source of short-lived radionuclides. He argues for stellar winds from nearby red giant stars within the molecular cloud parental to the solar nebula. Dearborn & Blake (1984, 1985) propose massive stars (greater than $100 M_{\odot}$) as a source of ^{26}Al for the Solar System and the ISM.

A striking observation in the meteoritic data is the rarity of examples in which radiogenic ^{26}Mg and large ^{50}Ti anomalies occur in the same sample (figure 7). In all but one case in which the ^{50}Ti anomaly exceeds 2 ‰, no radiogenic ^{26}Mg has been detected, even though Al/Mg ratios are high. This is especially noteworthy in the FUN inclusions, which are not distinguishable

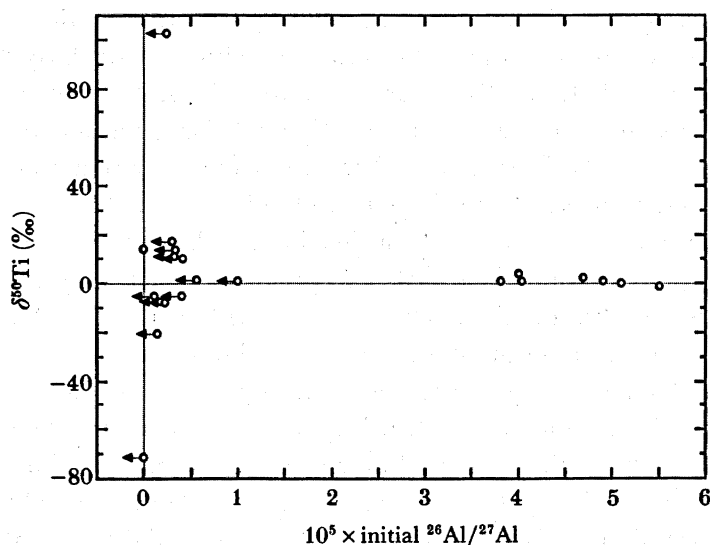


FIGURE 7. Samples with nucleosynthetic anomalies in ^{50}Ti show no evidence for radiogenic ^{26}Mg , and vice versa. ^{50}Ti and ^{26}Al are produced in different astrophysical settings, but the reason for the anticorrelation is unclear.

from other CAI on the basis of chemistry or mineralogy. The FUN inclusions lack radiogenic ^{26}Al . As noted by Hutcheon *et al.* (1986), CAI consisting only of hibonite usually have no ^{26}Mg excess and have ^{50}Ti anomalies; CAI in which hibonite is accompanied by spinel and/or melilite have ^{26}Mg excess and no ^{50}Ti anomaly. From the discussion above, it is very likely that the anomalies in neutron-rich isotopes originate in supernova sources, and the ^{26}Al originates in some other source or sources. It is not clear, however, why CAI possess one or the other of these effects, but not both.

4. OTHER EXTINCT RADIOACTIVITIES

Additional effects due to extinct radionuclides are expected in ^{53}Cr (from ^{53}Mn) and ^{60}Ni (from ^{60}Fe). Birck & Allègre (1985) found both excesses and deficiencies of ^{53}Cr in Allende CAI, with $^{53}\text{Cr}/^{52}\text{Cr}$ correlated with $^{55}\text{Mn}/^{52}\text{Cr}$ in an isochron diagram. The inferred initial $^{53}\text{Mn}/^{55}\text{Mn}$ ratio was about 4×10^{-5} . Lee (1987) showed that the interpretation of their data is not straightforward and requires multistage evolution, heterogeneous ^{53}Mn distribution, or intrinsic ^{53}Cr anomalies. Scatena-Wachel (1986) found excess ^{53}Cr in a Mn-rich carbonate in Orgueil and calculated an initial $^{53}\text{Mn}/^{55}\text{Mn} = 2.3 \times 10^{-7}$, implying either a 24 Ma interval between formation of Allende CAI and Orgueil carbonate, or a heterogeneous distribution of ^{53}Mn . Interpretation as a time interval is in conflict with ^{129}I – ^{129}Xe chronology of these meteorites (Lewis & Anders 1975; Wasserburg & Huneke 1979), so that isotopic heterogeneity appears to be preferred.

Iron-60, with a half-life of 1.5 Ma, decays via ^{60}Co to ^{60}Ni . No positive identification of radiogenic ^{60}Ni has yet been reported. Upper limits for $^{60}\text{Fe}/^{56}\text{Fe}$ on the order of 10^{-6} have been established in nickel-poor phases in iron meteorites (Scatena-Wachel *et al.* 1985).

Hutcheon *et al.* (1984) have sought evidence for excesses of ^{41}K due to decay of extinct ^{41}Ca ($t_{1/2} = 0.12$ Ma). In two inclusions containing radiogenic ^{26}Mg with inferred

$(^{26}\text{Al}/^{27}\text{Al})_0 \sim 5 \times 10^{-5}$, their data place an upper limit of $(8 \pm 3) \times 10^{-9}$ for $(^{41}\text{Ca}/^{40}\text{Ca})_0$. This low abundance of radiogenic ^{41}K requires a decay period of at least 1.8 Ma if the production ratio $^{41}\text{Ca}/^{40}\text{Ca}$ was $\sim 10^{-4}$.

5. CONCLUSIONS

Mass-dependent isotopic fractionation is observed for magnesium, silicon, calcium and titanium in those refractory components in primitive meteorites (such as CAI) which have undergone extensive high-temperature processing in the solar nebula. Such isotopic effects are small to non-existent in chondrules in ordinary chondrites, consistent with the lack of large fractionation of volatile from non-volatile elements in chondrules (Gooding *et al.* 1980; Grossman & Wasson 1982). The oxygen isotopic differences between ordinary chondrites and carbonaceous chondrites require separate reservoirs of precursor matter (Clayton *et al.* 1983a). These two systems must have been separated spatially or temporally in the solar nebula. The isotopic data imply that the material of carbonaceous chondrites underwent more intensive processing in the solar nebula, as well as the later aqueous alteration within the parent bodies of the C1 and C2 meteorites (Clayton & Mayeda 1984).

Oxygen isotopic heterogeneity in meteorites has been observed on all scales studied, from micrometers to planetesimal size. The nucleosynthetic anomalies in other rock-forming elements have so far been observed only in carbonaceous chondrites. It is not yet known whether this is a real difference between parts of the solar nebula, or whether it is an artefact of inadequate study of primitive ordinary chondrites. Much evidence suggests inhomogeneous distribution of short-lived radionuclides, particularly ^{26}Al , ^{53}Mn and ^{129}I , but such inhomogeneity has no necessary implications for the distribution of anomalies in stable nuclei, such as ^{48}Ca and ^{50}Ti .

Both carbonaceous chondrites and unequilibrated ordinary chondrites contain materials with large variations in D/H and $^{13}\text{C}/^{12}\text{C}$, which have been ascribed to preservation of presolar compounds (see, for example, Swart *et al.* 1983; Yang & Epstein 1983; Kerridge 1983). Large nucleosynthetic anomalies in the noble gases, especially xenon, are also preserved in both the carbonaceous chondrites and the unequilibrated ordinary chondrites (Lewis *et al.* 1975; Alaerts *et al.* 1979). Some of the anomalous gases are carried in diamond grains, a few nanometers in size, of probable presolar origin (Lewis *et al.* 1987). In most cases, the isotopic anomalies in hydrogen, carbon and noble gases are not observable by bulk analysis of the whole meteorite, but require techniques for resolving minor chemical components, such as stepped heating or acid demineralization, to reveal the anomalies. The ion microprobe has proved very effective in revealing very large isotopic anomalies in carbon and hydrogen in very small samples (McKeegan *et al.* 1985; Zinner & Epstein 1986). The same may be true for the rock-forming elements; perhaps isotopic study of the components of unequilibrated ordinary chondrites on a sub-micron scale may reveal a wealth of information. New analytical techniques must be developed to permit such studies.

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