



I-Xe measurements of CAIs and chondrules from the CV3 chondrites Mokoia and Vigarano

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Abstract—I-Xe analyses were carried out for chondrules and refractory inclusions from the two CV3 carbonaceous chondrites Mokoia and Vigarano (representing the oxidized and reduced subgroups, respectively). Although some degree of disturbance to the I-Xe system is evident in all of the samples, evidence is preserved of aqueous alteration of CAIs in Mokoia 1 Myr later than the I-Xe age of the Shallowater standard and of the alteration of a chondrule (V3) from Vigarano ~0.7 Myr later than Shallowater. Other chondrules in Mokoia and Vigarano experienced disturbance of the I-Xe system millions of years later and, in the case of one Vigarano chondrule (VS1), complete resetting of the I-Xe system after decay of essentially all ¹²⁹I, corresponding to an age more than 80 Myr after Shallowater. Our interpretation is that accretion and processing to form the Mokoia and Vigarano parent bodies must have continued for at least 4 Myr and 80 Myr, respectively. The late age of a chondrule that shows no evidence for any aqueous alteration or significant thermal processing after its formation leads us to postulate the existence of an energetic chondrule-forming mechanism at a time when nebular processes are not expected to be important.

INTRODUCTION

The aim of this study is to determine the timing of the alteration events in objects from CV3 carbonaceous chondrites by studying the I-Xe systematics within CAIs and chondrules from the meteorites Vigarano and Mokoia. Carbonaceous chondrites contain some of the best-preserved material surviving from the formation of the solar system: their bulk elemental compositions are close to solar values, indicating that little differentiation of material occurred before their formation. They generally consist of refractory calcium-aluminium-rich inclusions (CAIs) and chondrules embedded in a fine-grained matrix. The matrix material is dominated by silicates but also includes oxides, sulfides, sulfates, carbonates, metal, and carbonaceous material in an unequilibrated assemblage (Brearley and Jones 1998) inferred to be the result of various alteration processes that occurred either in the solar nebula or upon an asteroidal parent body.

The Vigarano-type carbonaceous chondrite group, most of the members of which are classified as petrographic type 3, is among the least aqueously altered types of carbonaceous chondrite. Nonetheless, the various components within the

meteorites have been subject to one or more episodes of alteration either in the solar nebula or later within a parent body, or perhaps in both environments (Krot et al. 1995; Bischoff 1998; Sears and Akridge 1998; Zolensky et al. 1997). Radiometric dating of the alteration events may allow greater insight into the nature of the environment in which alteration occurred (e.g., very late ages would imply a parent body location rather than nebular).

The CV3 meteorites can be sub-divided using opaque mineralogy (e.g., the proportion of metal to magnetite) and other criteria into three subgroups distinguished by the extent and style of alteration they have undergone (McSween 1977; Weisberg et al. 1997; Hoffman et al. 2000). The CV3 reduced subgroup (CV3_R), typified by a large fraction of the Vigarano breccia, has undergone only minor hydrous alteration resulting in the formation of small amounts of ferrihydrite and smectite within the matrix. This is inferred to have occurred in a low-temperature, oxidizing parent body environment (Lee et al. 1996). The Allende-like oxidized subgroup (CV3_{OxA}) shows extensive alteration and replacement of minerals indicative of the presence of an aqueous phase (Krot et al. 1995; Kimura and Ikeda 1998). The Bali-like oxidized subgroup (CV3_{OxB}) has less metal, a greater abundance of

matrix material, a greater abundance of hydrated low-calcium phyllosilicates, and a greater variation in matrix fayalitic olivine composition than the CV3_{OxA} subgroup. Mokoia is reported (Krot et al. 1998) to be a breccia containing both OxA and OxB lithologies.

Although these observations strongly suggest that all CV3 meteorites have undergone aqueous alteration on a parent body, it is possible that some alteration features observed within refractory material predate any parent body alteration and may be due to gas phase reactions in the nebula before accretion (e.g., Wark 1981; Cohen et al. 1983; Keller and Buseck 1991). Many of the CV meteorites are polymict breccias of the three subgroups, and several also contain dark inclusions, which are clasts of material that has been extensively altered and then thermally metamorphosed (Johnson et al. 1990).

PREVIOUS I-Xe STUDIES AND OTHER CHRONOMETERS

Previous I-Xe studies have resulted in the apparent observation of a very early I-Xe whole rock age for Vigarano, which has been interpreted as either representative of the formation age of CAIs within the rock (Crabb et al. 1982) or as indicative of early alteration introducing iodine (Swindle 1998; Swindle et al. 1998). Analysis of Mokoia gave a distinct whole rock initial iodine value (Crabb et al. 1982) corresponding to an age 3.3 Myr later than Vigarano but still much earlier than closure in the Shallowater aubrite ($^{129}\text{I}/^{127}\text{I} = 1.125 \times 10^{-4}$ [Hohenberg 1967]). However, the initial iodine value adopted for the reference standard (Murchison magnetite) for the work by Crabb et al. on Mokoia and Vigarano was probably incorrect (Hohenberg et al. 2000; Pravdivtseva et al. 2003b), requiring a revision of the whole rock ages reported for Vigarano and Mokoia to 10 Myr later. The resulting adjusted whole rock I-Xe ages would be more plausible, at 2.1 Myr later than Shallowater for Vigarano and 5.4 Myr later than Shallowater for Mokoia. The relatively high bulk ratio of $^{129}\text{Xe}^*/\text{I}$ in Vigarano, compared to the oxidized subgroup, and the lower iodine concentration (Crabb et al. 1982) can perhaps be understood as a consequence of less extensive (aqueous) alteration (Gilmour 2000).

Recent studies have demonstrated a range of I-Xe ages in objects from CV chondrites (Swindle 1998; Pravdivtseva et al. 2003), suggesting that the apparent whole rock ages are an artefact. In Allende, a member of the same subgroup as Mokoia, secondary sodalite has been shown to be the major host for iodine in CAIs, chondrules, and matrix (Zaikowski 1980; Swindle 1998; Kirschbaum 1988; Wasserburg and Huneke 1979), implying that the I-Xe chronometer is recording the alteration of these components. Dark inclusions in Allende have I-Xe ages reflecting alteration before incorporation in Allende, but the inclusions had already been incorporated into the rock when subsequent alteration some

4 Myr later set CAI ages (Pravdivtseva et al. 2003b; Krot et al. 2002). I-Xe dating of CAIs, chondrules, and dark inclusions from Efremovka (CV3_R) has shown a large range of apparent ages suggesting continuing alteration (Swindle 1998; Swindle et al. 1998; Krot et al. 1999). I-Xe dating of mineral separates from Bali and Kaba (both members of the Bali-like CV3_{OxB} subgroup) suggests that oxidation of metal and formation of phyllosilicates continued until at least 10 Myr after the formation of the enstatite in these meteorites but did not affect the I-Xe systematics of enstatite (Pravdivtseva et al. 2001).

Mn-Cr dating of fayalite in chondrules from Mokoia (Hutcheon et al. 1998) suggests that the fayalite formed in a single event between 7 and 16 Myr after Allende CAIs, in broad agreement with the I-Xe ages for the formation of magnetite and phyllosilicates in Kaba and Bali. (The range in ages reflects the choice of reference system rather than uncertainty in the data.) Hutcheon et al. favor an asteroidal rather than nebular environment for the formation of the fayalite on the grounds that the inferred initial $^{53}\text{Mn}/^{55}\text{Mn}$ values are rather similar to those found in differentiated meteorites. Al-Mg dating gives a range of ages for CAIs and chondrules from Axtell (CV3_{OxA}), suggesting an extended pre-compaction history (Srinivasan et al. 2000).

Thus, radiometric ages in conjunction with petrography and mineralogy lead to an inferred complex history recorded within CV meteorites of pre-accretionary events, regolith gardening, and aqueous and thermal processes on the parent body. In this study of separated components from Mokoia and Vigarano, our aim was to verify the recalibration of the Vigarano whole rock I-Xe isochron, to investigate what significance may be attached to the whole rock isochrons, and to test the hypothesis, based on whole rock $^{129}\text{Xe}/\text{I}$ ratios, that alteration was more prolonged in the CV3_{Ox} than in the CV3_R subgroup.

ANALYTICAL PROCEDURES AND DATA REDUCTION

The two meteorites chosen for this study, Vigarano and Mokoia, are both brecciated rocks. Vigarano, although predominately consisting of CV3_R material, includes clasts of both CV3_{OxA} and CV3_{OxB} material (Krot et al. 2000) as well as dark inclusions (Johnson et al. 1990). Mokoia includes Bali-like material, metamorphosed clasts (Krot and Hutcheon 1997), and dark inclusions (Ohnishi 2000). Portions of CAIs and chondrules were manually separated from specimens of Mokoia (BM1910,279) and Vigarano (BM1920,347) at the Natural History Museum, London, and broken into pieces. The Mokoia sub-sample was predominately of the CV3_{OxA} type, and the Vigarano specimen appeared to be representative of the reduced lithology. Part of each separated object was used for I-Xe analysis, and the remainder was polished for petrographic and mineralogical analysis. In some

cases, the material intended for I-Xe analysis was further subdivided before neutron irradiation, and each piece was analyzed separately. The decision as to whether or not subsamples were used for I-Xe measurements or petrography was essentially random—no attempt was made to select iodine-rich portions, and it is believed that the irradiated subsamples were reasonably representative of the objects from which they were taken. Polished chips for petrographic use were prepared without the use of water to avoid the loss of any potentially water-soluble minerals. Textural observations were made using images from both SEM and FEG-SEM instruments; descriptions and analyses of the samples are given below. Masses of the portions used for I-Xe analysis are shown in Table 1.

Samples for I-Xe analysis were neutron irradiated at the Ford reactor, Michigan, (our laboratory designation irradiation MN12) in the L67 water cooled position. Hornblende Hb3gr monitors (Turner 1971; Roddick 1983) indicated a neutron fluence of $3.3 \times 10^{18} \text{ cm}^{-2}$ fast and $8.2 \times 10^{18} \text{ cm}^{-2}$ thermal. Aliquots of a non-magnetic separate from the enstatite achondrite Shallowater were included in the irradiation as an iodine conversion standard. Xenon isotopic analysis of the irradiated samples was carried out using the RELAX resonance ionization mass spectrometer (Gilmour et al. 1994) in conjunction with laser-stepped heating. This allows a lower blank than a conventional furnace, but accurate temperatures are not available. Using an initial $^{129}\text{I}/^{127}\text{I}$ value for Shallowater of 1.125×10^{-4} (Hohenberg 1967), a ^{127}I to ^{128}Xe conversion efficiency of 7.6×10^{-5} was obtained with an internal uncertainty of 1.5% derived from an isochron diagram for six aliquots of the standard.

The cold blanks during the period over which these measurements were carried out were $\sim 1 \times 10^{-16} \text{ cm}^3 \text{ STP } ^{132}\text{Xe}$ and approximately atmospheric in composition; blank corrections usually accounted for less than 10% of ^{132}Xe measured in a single step. When assessing I-Xe systematics, formal corrections were made for xenon due to induced

fission on the basis of a trapped component (assumed to be Q) and the measured $^{134}\text{Xe}/^{132}\text{Xe}$ ratio. No correction was made for spallation. Age spectra, except where otherwise noted, are plotted with the assumption that the trapped $^{129}\text{Xe}/^{132}\text{Xe}$ value was that of Q-xenon (Busemann et al. 2000). The error bars in all cases represent an estimate of one standard deviation (disregarding systematic errors due to, e.g., uncertainty in calibration aliquots) and do not include any contribution from the uncertainty in end members used during data reduction. Concentrations of uranium and barium and/or tellurium were estimated from the over-abundance of ^{134}Xe from induced fission or ^{131}Xe from the reactions $^{130}\text{Ba}(n, e\text{-capture})^{131}\text{Xe}$ and $^{130}\text{Te}(n, 2\beta)^{131}\text{Xe}$ together with the neutron fluences given above and published cross-sections, branching ratios, and fission yields (General Electric 1988; Ozima and Podosek 2002).

Xenon isotope data for all samples, corrected only for instrumental effects and blanks, are available either as an electronic supplement from the journal Web site or from the corresponding author.

MINERALOGY/PETROGRAPHY

One CAI and 5 chondrules from Vigarano and two CAIs and two chondrules from Mokoia were studied. For chondrules V3 and V4, two aliquots were irradiated. Backscattered electron images are shown in Figs. 1 and 2, and compositions and textures are summarized here.

VS1

Vigarano VS1 is a large ($\sim 5 \text{ mm}$) chondrule composed of stubby to skeletal enstatite laths and rounded olivine phenocrysts in a mesostasis composed of feldspar and silicon-rich glass. Diopside and augite are present as minor phases. The chondrule contains blebs of Fe-Ni metal and is cross-cut by iron-rich veins.

Table 1. Summary of data from all analyzed samples.^a

Sample	VS1	V1	V2	V3a	V3b	V4a	V4b	V5	M1	M2	M3	M4
Type	chondrule	chondrule	chondrule	chondrule	chondrule	chondrule	chondrule	CAI	CAI	CAI	chondrule	chondrule
Mass (μg)	490	767	6685	4087	357	1460	500	873	1695	884	1243	1272
^{127}I (ppb)	2	19	18	33	30	14	43	42	131	141	45	18
^{132}Xe (10^{-12} ccSTP/g)	4.1	10.2	7.7	44.2	16.5	6.3	61.7	1800	53.8	128.0	63.0	12.3
U (ppb)	2.9	18	4.9	8.1	18	15	48	74	24	5.8	37	12
Ba (ppm)	0.2	0.2	1.2	1.7	2.2	8.9	25	16	8.4	8.2	0.7	2.5
Te (ppb)	20	22	130	190	240	980	2700	170	930	900	80	270
I/Xe atomic (10^4)	2.9	8.8	11	3.6	8.5	11	3.3	0.11	12	5.2	3.4	6.8
"Age" (Myr)	reset late	~ 3	~ 3	0.7	0.7	–	–	–	~ 1	~ 1	> 5	> 5

^aThe mass refers to the sample split used for I-Xe measurements and not to the original mass of the object. Barium and tellurium concentrations are upper limits calculated from ^{131}Xe excesses due to neutron capture during the artificial irradiation, assuming, respectively, that either only barium or only tellurium was present. See text for description of calculation of U and Ba/Te concentrations. External uncertainties for noble gas concentrations are $\sim 20\%$. Ages are given in millions of years after closure of the I-Xe system in Shallowater, however most samples showed complex I-Xe systematics so these numbers are only a guide—see text for discussion.

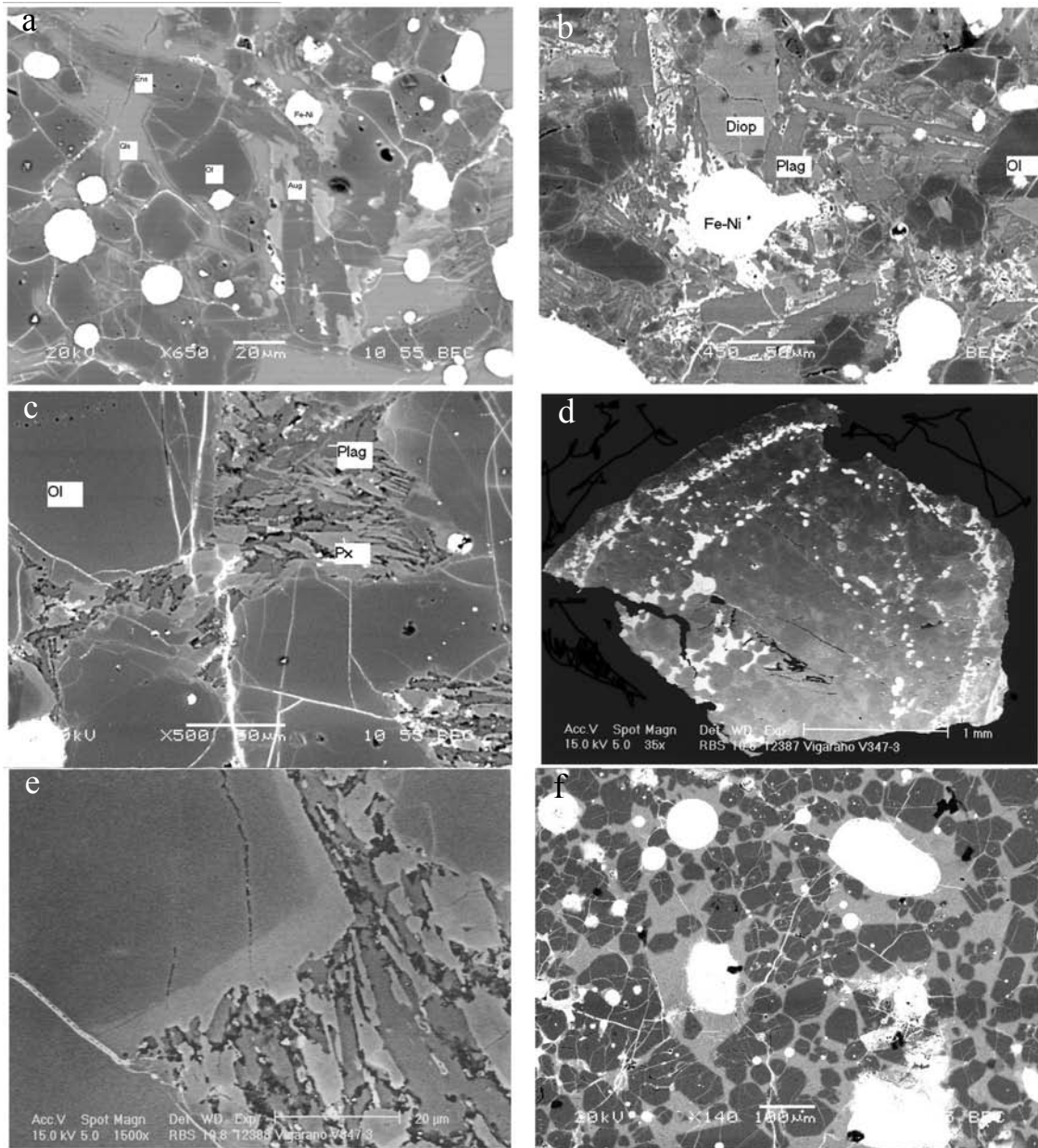


Fig. 1. Backscattered electron images of samples (see text for full descriptions). From top-left to right: a) chondrule VS1; b) chondrule V1; c) chondrule V2 showing corrosion along plagioclase grain boundaries; d) chondrule V3; e) enlargement of V3 showing iron-rich rim on olivine crystal and alteration of mesostasis to sodalite; f) chondrule V4—no alteration is apparent.

V1

Vigarano V1 is an aluminium-rich chondrule composed of forsteritic phenocrysts surrounded by plagioclase laths and interstitial diopside. The chondrule contains large Fe-Ni and FeS blebs, up to 200 μm across.

V2

Vigarano V2 is a large (2 mm in diameter) porphyritic olivine chondrule. Large rounded olivine grains are set in a

diopside and plagioclase mesostasis. The plagioclase appears to have reacted along grain boundaries to form an iron-poor phase too small to analyze with the electron probe. X-ray mapping showed minor chlorine associated with the diopside mesostasis. Metal blebs have a complex rim, possibly due to corrosion.

V3

Vigarano V3 is also a large (2 mm in diameter) porphyritic olivine chondrule. The olivines have an iron-rich

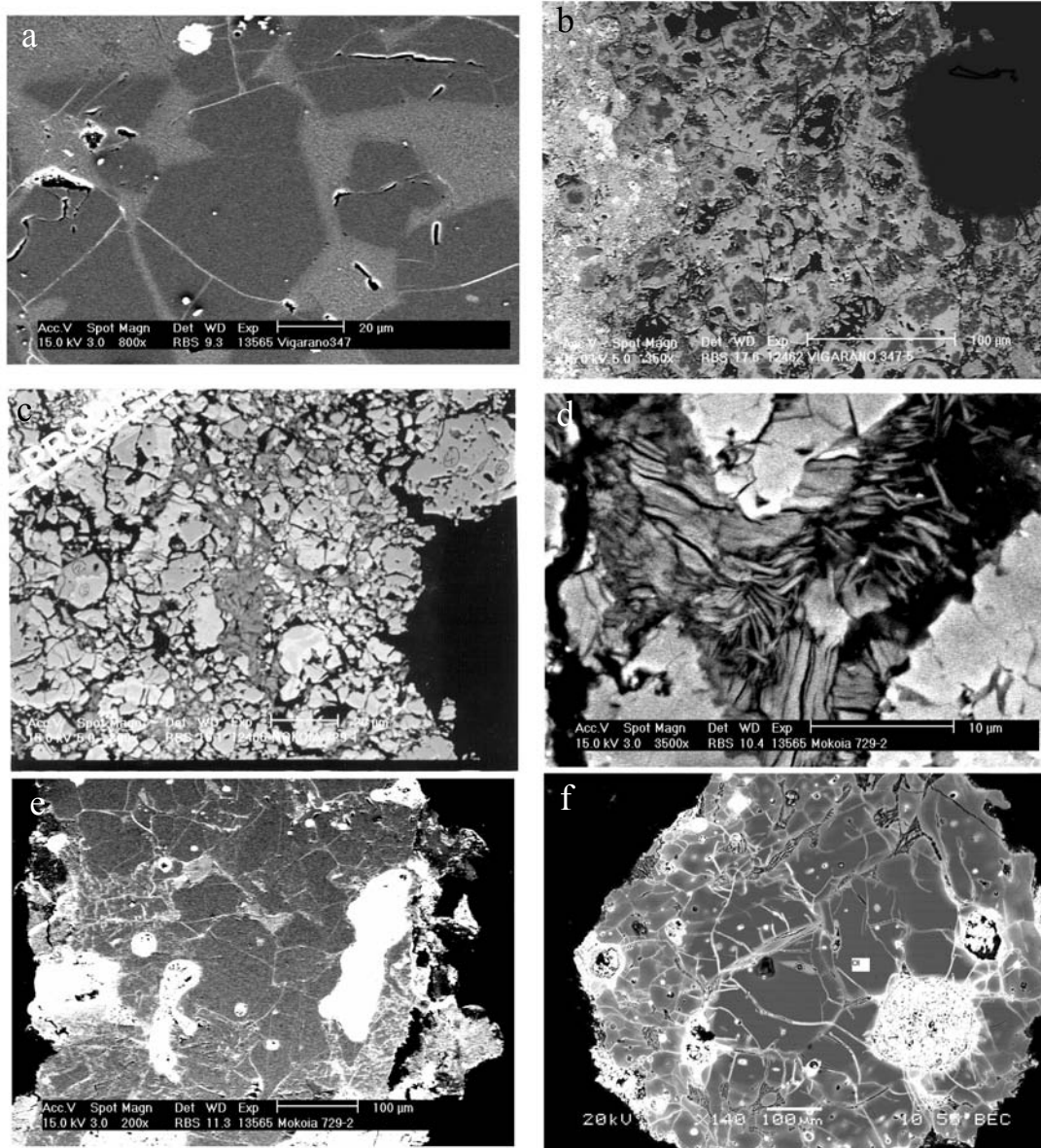


Fig. 2. Backscattered electron images of further samples (see also Fig. 1). From top-left to right: a) enlargement of chondrule V4 illustrating absence of alteration along grain boundaries; b) unaltered Vigarano CAI V5; c) Mokoia CAI M1 showing extensive alteration to sodalite and a Na, Al-rich phyllosilicate; d) high magnification image of the similar Mokoia CAI M2 showing alteration to a phyllosilicate; e) chondrule M3; f) chondrule M4.

rim. The diopside and plagioclase mesostasis has been partly altered to sodalite. The chondrule contains blebs of Fe-Ni metal and FeS.

V4

Vigarano V4 is a porphyritic chondrule composed of olivine crystals, $Fo_{98.3-99.4}$, typically 50–100 μm across, surrounded by a mesostasis composed of a devitrified Ca, Al-rich glass and submicron blebs of diopside. The chondrule contains numerous blebs, up to several hundred microns across, of FeS and Fe-Ni metal.

V5

Vigarano V5 is a fine-grained CAI consisting of åkermanitic melilite nodules enclosing spinel cores. The spinel contains minor lath shaped hibonite crystals and numerous submicron perovskites. Many of the nodules are surrounded by rims of anorthite with an outer Al-rich diopside layer.

M1

Mokoia M1 is a fine-grained CAI exhibiting extensive alteration. Some primary nodules of spinel surrounded by

diopside are present, but in most parts of the CAI, these primary minerals have been replaced by sodalite and a phyllosilicate rich in sodium and aluminium.

M2

Mokoia M2 is a fine-grained CAI. It is composed of nodules of spinel (~10–50 μm across) with a variable composition containing up to 6 wt% FeO. The spinel nodules are surrounded by rims of Al, Ti-rich clinopyroxene. These patches of primary minerals are enclosed within abundant sodalite, andradite, hedenbergite, and phyllosilicates.

M3

Mokoia 729-3 is a pyroxene-olivine porphyritic chondrule. The olivine (Fo_{98}) grains tend to be rounded in shape; enstatite ($\text{Fs}_{1.8}$; $\text{Wo}_{1.1}$) is abundant, sometimes enclosing olivine. The pyroxene is often cracked along its cleavage planes with an iron oxide filling the cracks. The chondrule contains abundant sulphide blebs. The mesostasis is composed of Al-rich diopside and rare glass.

M4

Mokoia M4 is a porphyritic olivine chondrule composed of densely packed, rounded olivine grains. Olivine is highly forsteritic in the center of the chondrule (Fo_{97-99}) but more Fe-rich in the outer parts (Fo_{80-95}). The olivines are enclosed in a mesostasis of Al-rich diopside and partly devitrified silica-rich glass. The chondrule also contains several blebs, up to 100 μm across, of FeS and a calcium phosphate.

The CAIs M1 and M2 from Mokoia have clearly undergone alteration by an aqueous fluid subsequent to their formation, but the chondrules from Mokoia do not show any significant alteration. These observations are consistent with those of Cohen et al. (1983), who only reported phyllosilicates in refractory inclusions from Mokoia and found no evidence for aqueous alteration of the chondrules. However, Jones and Schilk (2000) have concluded that chondrules from Mokoia have generally experienced varying degrees of minor aqueous alteration resulting in, e.g., the presence of phyllosilicates in mesostasis but no alteration of olivine grains and little change in bulk chemical compositions. The chondrules from Vigarano have been subject to some observable alteration, resulting in the formation of sodalite in V3 and the reaction of plagioclase and metal in V2. The CAI V5 from Vigarano shows no sign of any alteration subsequent to its formation.

The more extensive alteration of the CAIs in Mokoia compared to the chondrules might suggest that the CAIs were altered prior to incorporation. Alternatively, if alteration in Mokoia occurred after compaction in a parent body, then the much greater extent of alteration of the CAIs might be due to their finer grain size and the possible lower stability of the

refractory minerals relative to the chondrule silicates under the prevailing conditions. The lack of any alteration in the fine-grained CAI V5, despite the reaction of some Vigarano chondrules, suggests that the alteration of the chondrules in Vigarano must have taken place before final assembly of the rock.

I-Xe RESULTS

Iodine-xenon data can be represented graphically in several ways. When seeking evidence of mixing between two components, a three-isotope plot with a common denominator on each axis is normally used. A special case of this is the isochron diagram, when the two components are trapped xenon and iodine-derived xenon with, ideally, a single initial iodine ratio. This has been conventionally plotted as $^{129}\text{Xe}/^{132}\text{Xe}$ versus $^{128}\text{Xe}/^{132}\text{Xe}$ or $I/^{132}\text{Xe}$ (^{130}Xe may also be used in the denominator). In this representation, one end member (the iodine-derived component) plots inconveniently at infinity, and when the largest uncertainty is in the measurement of the normalising isotope, then the resulting highly correlated error-bars can obscure details of the plot. An alternative mathematically equivalent representation of the same information is to use $^{132}\text{Xe}/^{129}\text{Xe}$ versus $^{128}\text{Xe}/^{129}\text{Xe}$ (or $I/^{129}\text{Xe}$) in which all end members plot on the diagram. (The “age” information is now shown not by the gradient of any correlation line but, rather, by its intercept with the x-axis). Another way of assessing the data is to plot an age-spectrum, as widely used in Ar-Ar dating. This latter approach is particularly useful when both $^{129}\text{Xe}/^{132}\text{Xe}$ and $^{128}\text{Xe}/^{132}\text{Xe}$ ratios are high, thus making the figure insensitive to assumptions about the composition of the trapped component, which must be subtracted. Where a trapped component has been subtracted, we denote the excess over Q-xenon (unless otherwise noted) with a subscript as, e.g., $^{129}\text{Xe}_{\text{XS}}$. An age-spectrum plot has the advantage that the thermal behavior of the system is easier to see than in an isochron plot. We will make use of each of these representations as appropriate.

Results from xenon isotopic analysis of our samples are summarized in Table 1 and Fig. 3. It can be seen immediately that the CAI V5 is exceptionally rich in trapped xenon and that the Mokoia CAIs M1 and M2 have the highest iodine concentrations. Figures 4 and 5 summarize the I-Xe results of 3-isotope plots for all the samples from Mokoia and Vigarano, respectively. Each sample is discussed separately below.

Mokoia CAIs

M1 and M2 both show evidence of aqueous alteration. Their high total iodine abundance is consistent with the observation of copious secondary sodalite, which has been shown to be a significant host for iodine (e.g., Wasserburg and Huneke 1979; Kirschbaum 1988). The three-isotope summary plot in Fig. 4 shows some evidence in the high temperature

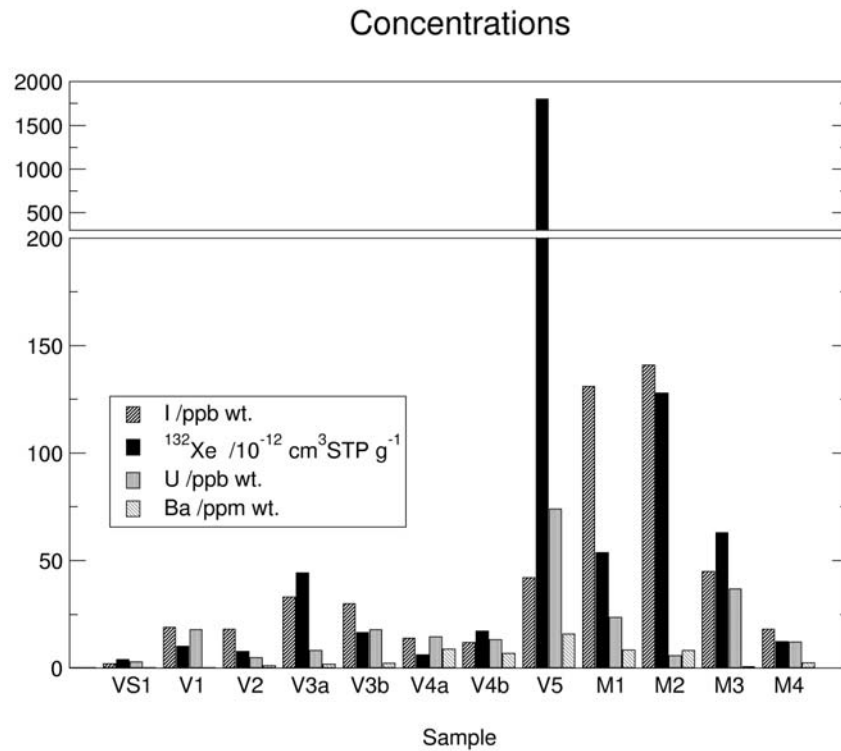


Fig. 3. A graphical representation of the concentrations from Table 1. Note the exceptionally high concentration of xenon in Vigarano CAI V5 and the high iodine concentrations in Mokoia CAIs M1 and M2.

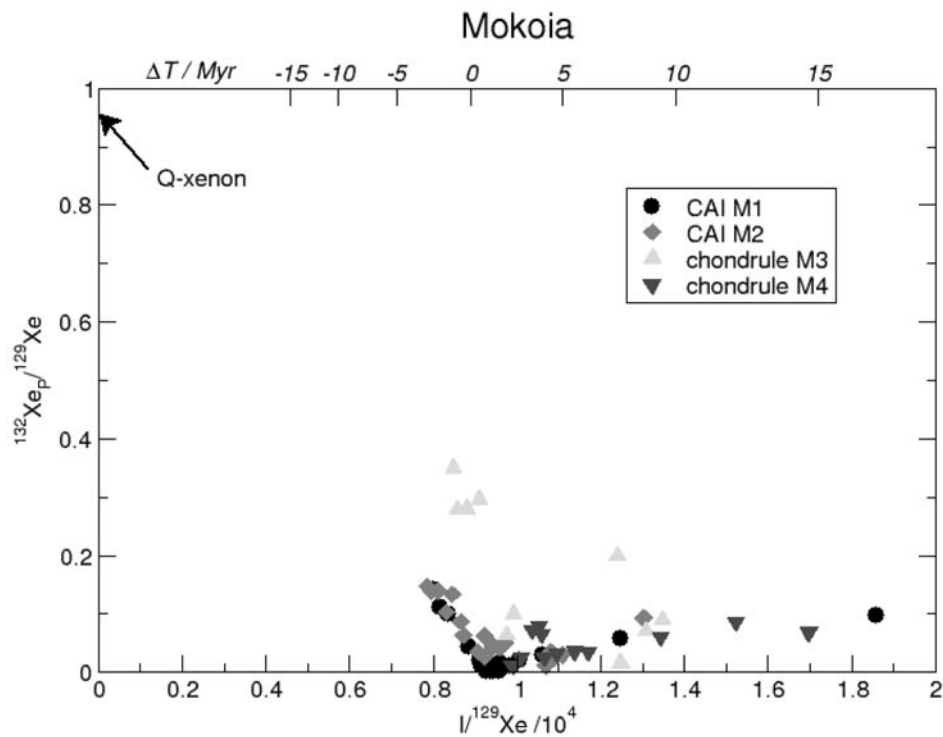


Fig. 4. Mokoia three-isotope summary plot; error bars have been suppressed for clarity, but symbol sizes are comparable to 1σ statistical errors in most cases. A few low-temperature releases with large amounts of iodine have been omitted. On this diagram, a pure xenon component plots on the y-axis, and a pure iodine component plots on the x-axis (later ages plot further to the right). Data from the heavily altered CAIs would scatter around a hypothetical mixing line between Q-xenon and an iodine end member with an initial $^{129}\text{I}/^{127}\text{I}$ value close to that of Shallowater.

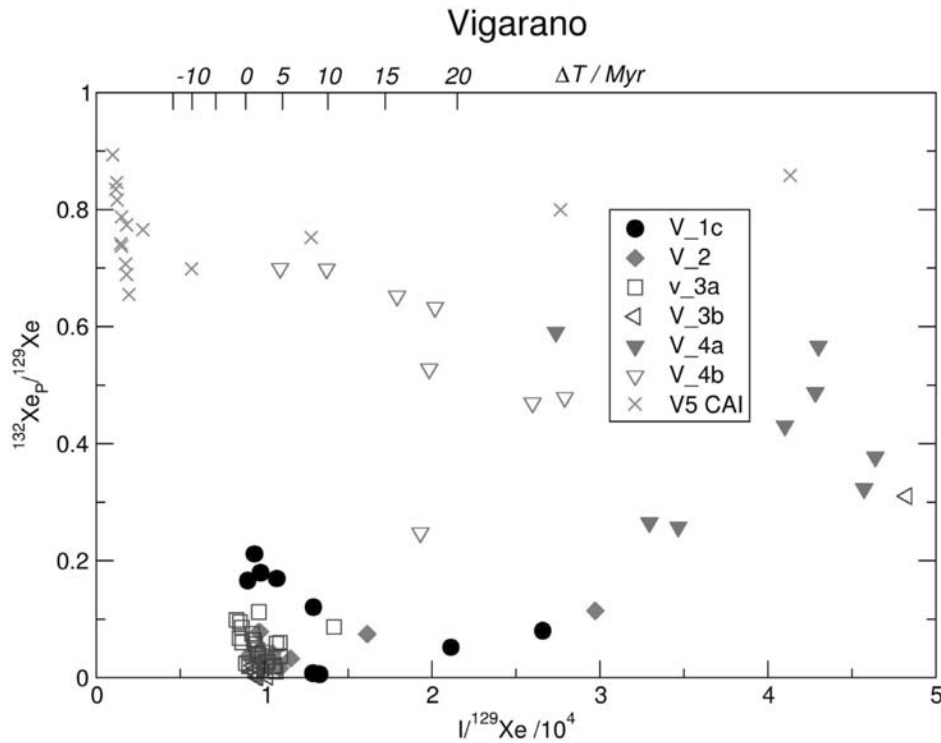


Fig. 5. Vigarano summary plot (chondrule VS1 not shown). Error bars have been suppressed for clarity, but symbol sizes are comparable to 1σ statistical errors in most cases. A few releases with large amounts of iodine have been omitted. The data points from the refractory inclusion V5 are clearly distinct from the chondrule data. With the exception of V4, the chondrule data (V1, V2, V3, V4) tends to cluster close to an iodine-rich end member with a $^{127}\text{I}/^{129}\text{Xe}$ value similar to that observed in the Shallowater enstatite achondrite I-Xe standard.

data for mixing between a trapped xenon component and an iodine-rich component with $I/^{129}\text{Xe}$ of about 9500, that is an initial $^{129}\text{I}/^{127}\text{I}$ ratio of about 1.05×10^{-4} . This initial iodine ratio corresponds to an age ~ 1 Myr later than Shallowater. However, there is scatter in excess of the estimated analytical errors, implying that the iodine-xenon system in these two objects has been subject to some disturbance. This can be seen more clearly in the age spectra shown in Fig. 6. The highest temperature releases from the two CAIs (comprising 10% of the iodine released from M1) give model ages consistent with each other. Both samples have their major iodine releases at intermediate temperatures accompanied by $^{129}\text{Xe}^*/\text{I}$ ratios lower than the maximum. M2 contains a greater concentration of trapped xenon than M1, possibly suggesting more extensive alteration of the former to fine-grained products. The later model ages for earlier release steps of M2 would also be consistent with suggestions that later I-Xe ages in Efremovka are associated with more extensive alteration (Krot et al. 1999).

Mokoia Chondrules

Chondrules M3 and M4 show no consistent ratio of $^{129}\text{Xe}^*$ and I. Model ages for each temperature step (shown in Fig. 7) are always later than those calculated for the high temperature releases from the CAIs M1 and M2 and are also

somewhat later than most, but not all, chondrule I-Xe formation ages (Whitby et al. 2002; Swindle et al. 1996). This suggests the possibility that there is no record of the I-Xe formation age of these chondrules but, rather, that the I-Xe system has been reset and disturbed at least 4 Myr after the formation of the sodalite in M1 and M2. (We note that the M4 data in Fig. 4 appear to trend systematically away from the CAI “intercept-age” in the direction of a component consisting of more recent iodine with admixed xenon.) Both M3 and M4 exhibited relatively large releases of trapped and ^{127}I -derived xenon from the lowest temperature step ($< 600^\circ\text{C}$), accounting for about 50% of the total iodine. This large release of trapped xenon at low temperature suggests a loosely bound, possibly surface site and may be due to the presence of a fine-grained alteration product such as the devitrified glass observed in M4.

Vigarano Chondrules

The Vigarano chondrule VS1 showed no significant ^{129}Xe excess over a trapped planetary xenon component and contained little total iodine or xenon, suggesting that this chondrule has partially lost some of its volatile elements. It is difficult to constrain when such a loss of volatile elements took place, but if all the iodine measured is intrinsic to the sample and there is no terrestrial contamination, then a lower

Age spectra for Mokoia CAIs M1 and M2

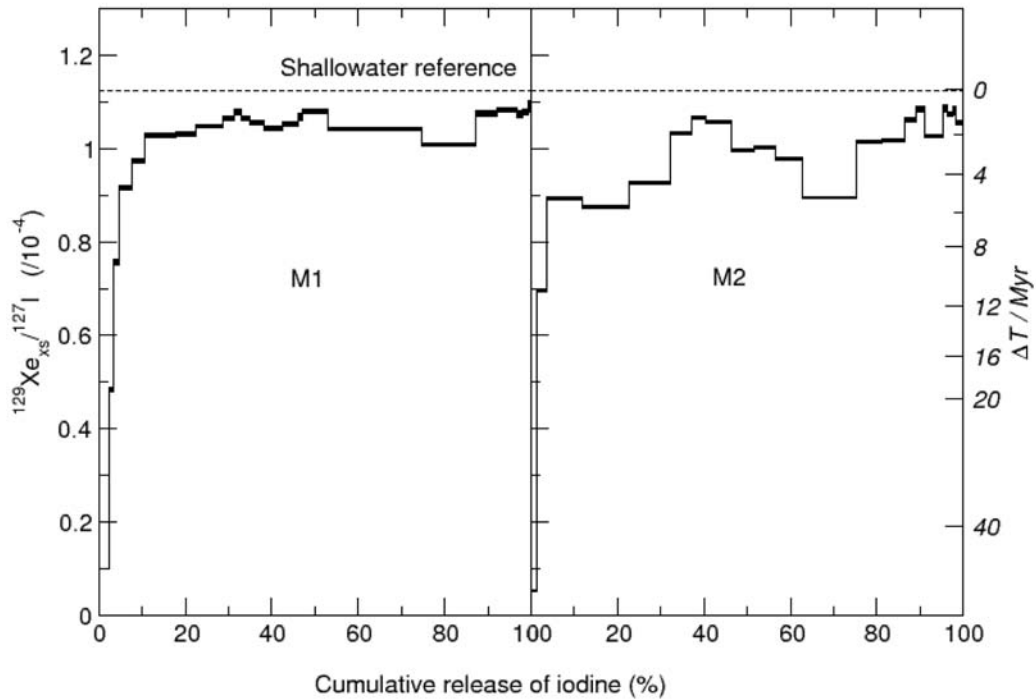


Fig. 6. Age spectra for the Mokoia CAIs. The right-hand axis shows age relative to the Shallowwater standard (positive values are later).

Age spectra for Mokoia chondrules M3 and M4

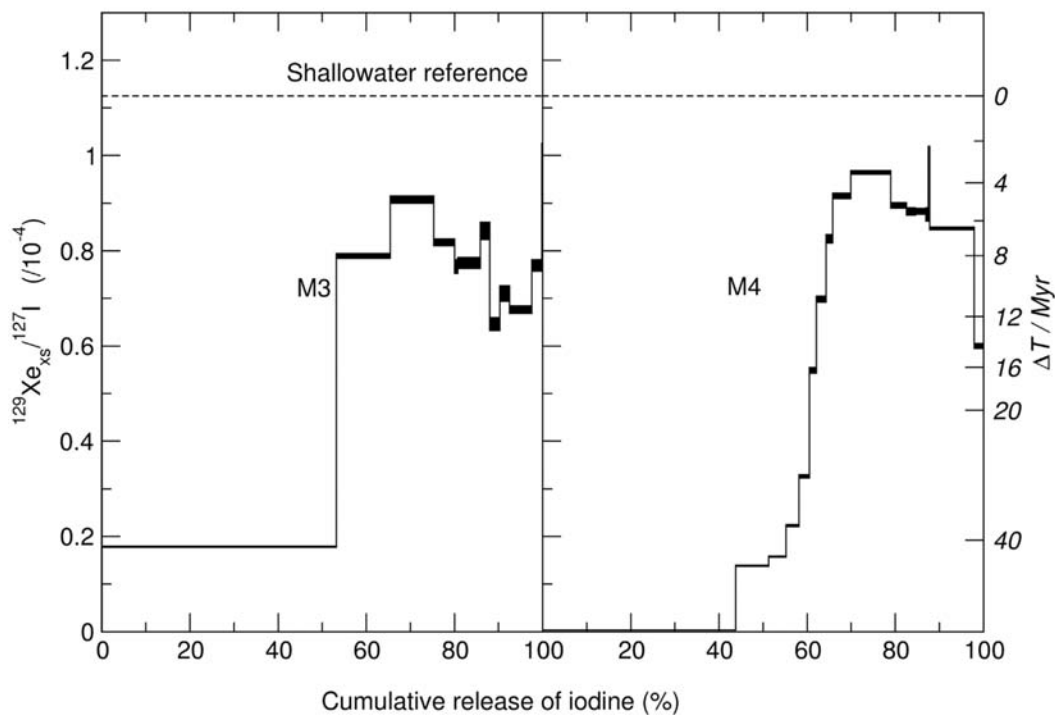


Fig. 7. Age spectra for chondrules M3 and M4. The right-hand axis shows age relative to the Shallowwater standard (positive values are later).

bound of at least 80 Myr later than closure of the I-Xe system in Shallowater can be calculated from the nominal ^{129}Xe excess over Q xenon plus three times the estimated error. Although contamination with terrestrial iodine can affect I-Xe systematics, giving apparently “late” ages, we feel this does not explain the data from VS1, as the low concentrations of iodine and xenon together with the presence of glass suggest that the sample has suffered only very minor weathering. VS1 was a large (~5 mm) chondrule, and it is not clear that “macrochondrules” formed in the same way as their smaller brethren, despite the textural and mineralogical similarities (Weisberg et al. 1988; Hutchison and Bridges 1995). Previous I-Xe experiments on other unusually large chondrules have also found very late closure ages (Gilmour et al. 1995, 2000). The trapped ^{132}Xe xenon concentration in VS1 of 4.1×10^{-12} $\text{cm}^3\text{STP/g}$ (some of which is probably due to terrestrial atmosphere) is lower than the values of 1.7×10^{-11} and 1.5×10^{-10} $\text{cm}^3\text{STP/g}$ for macrochondrules from Isoulane-n-Amahar (L6) and Parnallee (LL3), respectively (previously unpublished concentrations from the study of Gilmour et al. [2000]), and is also lower than the observed range for normalized chondrules of 2×10^{-11} to 2.5×10^{-10} $\text{cm}^3\text{STP/g}$. The low volatile concentrations and the late age are suggestive of an impact origin on an airless body or of volatile loss by some other mechanism after initial formation of the chondrule.

Figure 5 summarizes the I-Xe data for the other objects

from Vigarano. The high-temperature data for chondrules V1, V2 and V3 scatter together on a three-isotope plot with $I/^{129}\text{Xe}$ values of about 1×10^4 and so model initial $^{129}\text{I}/^{127}\text{I}$ ratios close to 1×10^{-4} . In contrast, V4 has clearly been disturbed later, and most releases from this chondrule have higher $^{132}\text{XeP}/^{129}\text{Xe}$ ratios than those observed for the other chondrules in this study. The iodine concentrations in V4 are similar to chondrules V1, V2, and V3 from Vigarano, so it is not the case that a common alteration event has simply had more effect on V4.

Figure 8 shows an isochron diagram for chondrule V2 in which there appears to be evidence of two events having been recorded by the I-Xe system. The four lowest temperature releases show no correlation, but the next five steps lie on a line corresponding to an age 50 Myr later than Shallowater. The twelve highest temperature steps define a line corresponding to an age 5 Myr later than Shallowater. Four intermediate temperature steps plot between the two lines. If the I-Xe system in this chondrule has recorded two distinct events, then some evidence of this should be seen in the mineralogy inferred from the xenon release pattern. Figure 9 shows both an age-spectrum representation of the same data and also a plot of $^{134}\text{Xe}_{\text{XS}}/^{131}\text{Xe}_{\text{XS}}$ as a proxy for the ratio of uranium (via induced fission) to either barium or tellurium (via neutron capture and subsequent decay). The age-spectrum representation (calculated using the trapped $^{129}\text{Xe}/$

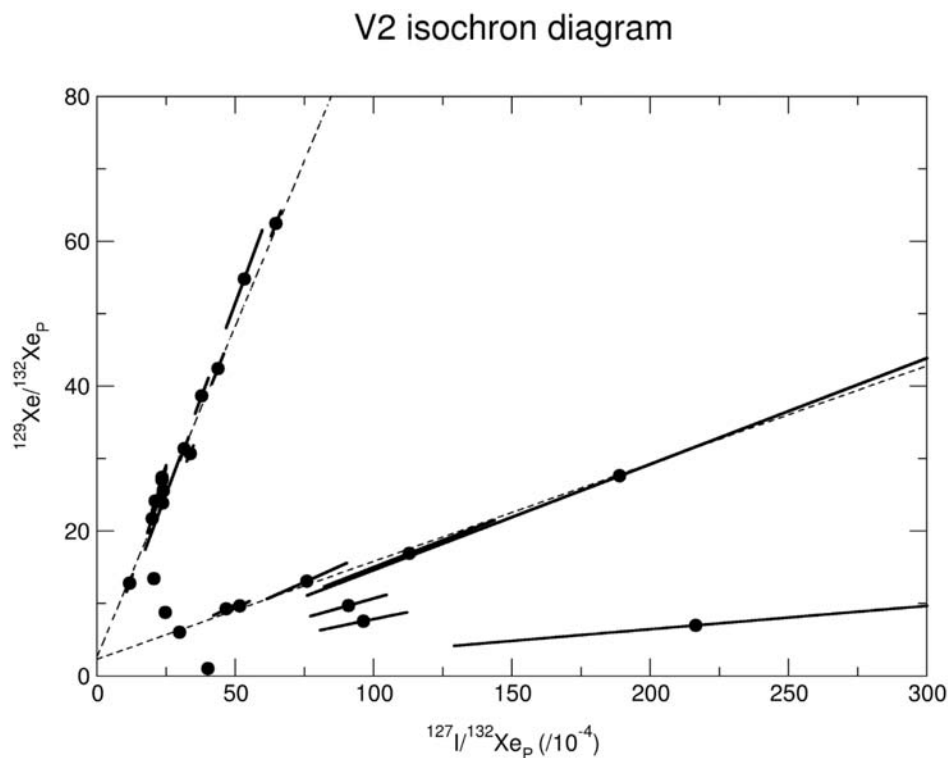


Fig. 8. Isochron diagram for Vigarano chondrule V2 showing I-Xe correlations for both high temperature releases and lower temperature releases. The dashed lines are fits to selected consecutive releases and correspond to ages of 5 and 50 Myr later than Shallowater. See discussion in text.

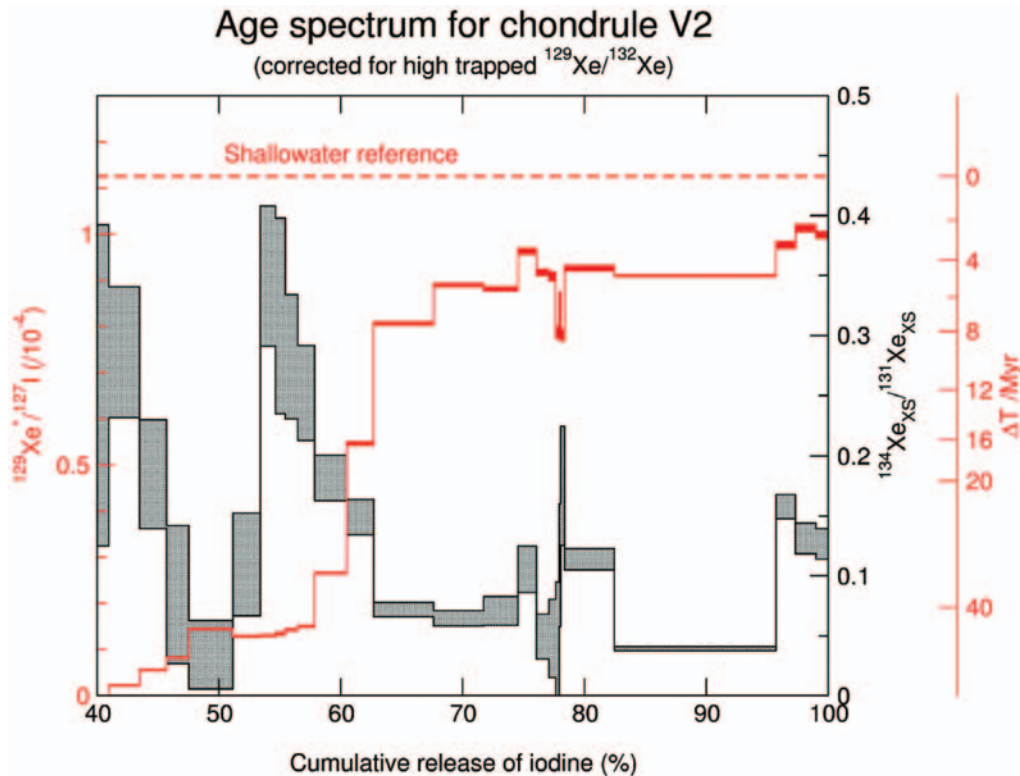


Fig. 9. Age spectrum for chondrule V2, also showing the ratio of excess ^{134}Xe (from induced fission of uranium) to excess ^{131}Xe (from neutron capture on barium and tellurium)—the excesses are denoted on the diagram as $^{134}\text{Xe}_{\text{XS}}$ and $^{131}\text{Xe}_{\text{XS}}$ and are calculated by subtracting the calculated trapped Q-component from the measured values. The age plateau between 48% and 58% of iodine released coincides with a peak in the the $^{134}\text{Xe}_{\text{XS}}/^{131}\text{Xe}_{\text{XS}}$ value, suggesting that a discrete mineral can be associated with the event recorded by this age—see discussion in text.

^{132}Xe values inferred from the isochron diagram) makes clearer the extent of scatter in the model ages than does the isochron diagram. Nonetheless, the interpretation of two I-Xe ages is supported by the peak in $^{134}\text{Xe}_{\text{XS}}/^{131}\text{Xe}_{\text{XS}}$ that is more or less coincident with the releases defining the later of the two ages (from 48% to 58% of the total iodine released). A plot of $^{131}\text{Xe}_{\text{XS}}/^{127}\text{I}$ (not shown) also supports the existence of a change in the mineralogy of the xenon host that is coincident with the two groups of data visible in the isochron diagram. The petrographic study of V2 revealed evidence of reaction along grain boundaries, and so the two events controlling the I-Xe systematics within this chondrule are plausibly interpreted as alteration 50 Myr later than Shallowater and a disturbed record of chondrule formation 5 Myr later than Shallowater. (We note that in the ordinary chondrite St. Séverin [LL6], uranium is associated with grain boundaries due to metamorphism [Jones and Burnett 1979]—this is consistent with our observations.) The inferred trapped $^{129}\text{Xe}/^{132}\text{Xe}$ values are 3.6 and 2.2 for the early and late ages respectively, so the changes in the trapped xenon composition cannot be due solely to evolution as a closed system. Given the petrographic evidence of an external agent, such closed system behavior would not be expected. (If the three highest temperature points are omitted from the data set, then the inferred trapped compositions become identical within error.)

That the values are higher than for primordial xenon ($^{129}\text{Xe}/^{132}\text{Xe} = 1.04$ for Q xenon) presumably reflects the high I/Xe ratios both within the material that formed the chondrule and in the fluid or vapor that caused the subsequent alteration.

Figure 10 shows age spectra for two sub-samples of chondrule V3, each showing similar behavior. The high temperature data from V3b define a plateau corresponding to an initial iodine ratio of $1.09 \pm 0.01 \times 10^{-4}$, or 0.7 Myr later than closure in Shallowater. V3a, the larger of the two sub-samples and for which smaller heating increments were used, shows an age spectrum generally increasing in age with temperature. The mean of the ten highest temperature releases from V3a (corresponding to 21% of the total ^{127}I) defines an initial iodine value indistinguishable from that of V3b. The two sub-samples do show differences in their concentrations of the $^{129}\text{Xe}^*$ (^{129}I) and $^{132}\text{Xe}_p$ components, both of which are lower in V3a than in V3b by a factor of three, although the total iodine concentrations are similar. Chondrule V3 shows clear evidence of secondary alteration, and it is likely that the I-Xe chronometer is recording the formation of sodalite from the mesostasis within the chondrule, presumably as a result of aqueous alteration; this alteration may also have been responsible for the formation of the iron-rich rims on the olivines. An alternative explanation, that the I-Xe age could be that of formation of the chondrule if the sodalite formed so

Age spectra for chondrule V3

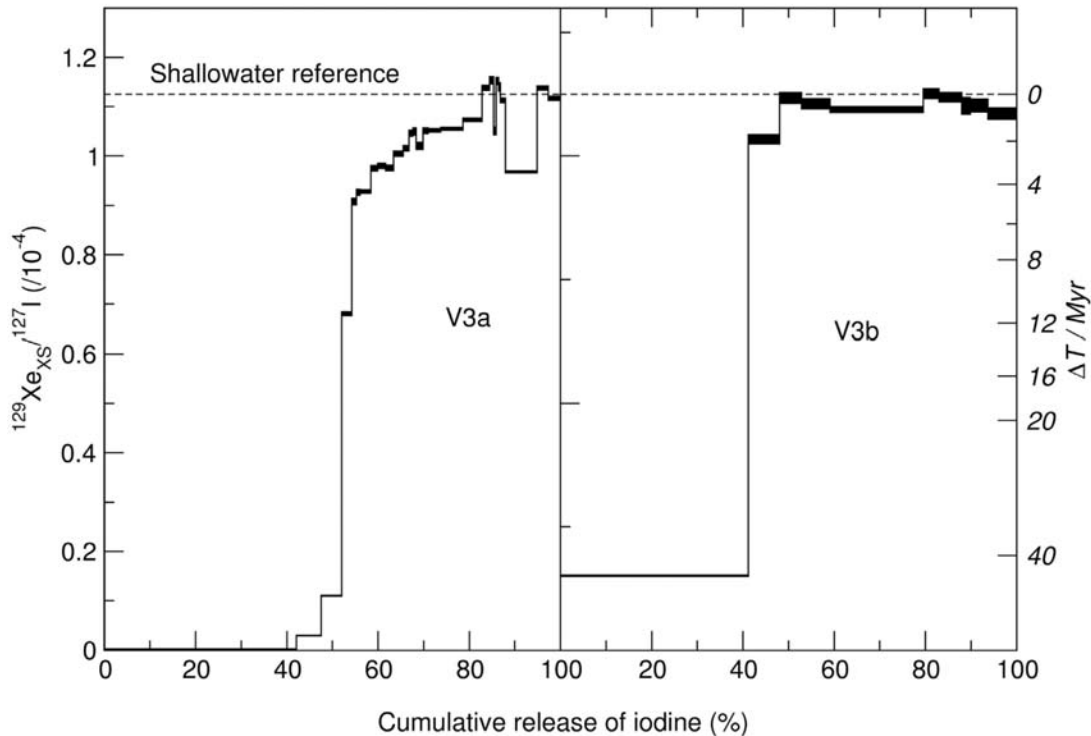


Fig. 10. Age spectra for two pieces of the Vigarano chondrule V3. High temperature data define an I-Xe age 0.7 Myr later than Shallowater.

much later that negligible ^{129}I was available, is less likely because sodalite, where present in chondrules, is usually the major iodine host.

Figure 5 shows clearly that chondrule V4 has higher $^{132}\text{Xe}_p/^{129}\text{Xe}$ ratios and a much greater range of I/ ^{129}Xe ratios than the other Vigarano chondrules. (In this respect, chondrule V4 appears more like the refractory inclusion V5 but has much lower absolute concentrations of iodine and trapped xenon.) Model initial iodine values for releases from V4 suggest a disturbance of the I-Xe system at least tens of million years later than in V1–V3. Although the two subsamples V4a and V4b had slightly different release patterns and trace element concentrations, presumably due to the heterogeneous distribution of minerals and glass within the chondrule, the I-Xe systematics are essentially indistinguishable. The mineralogy of V4 shows no clear evidence for aqueous alteration of the chondrule, and so it is more likely that the disturbance to the I-Xe system was caused by a thermal or mild shock event.

Vigarano CAI

Although apparently unaltered, the Vigarano CAI V5 had high concentrations of xenon and iodine, neither of which would be expected for the high-temperature, low-pressure nebular formation conditions usually ascribed to CAIs. The

high-temperature iodine-xenon data form a linear array on a three-isotope diagram demonstrating two-component mixing (see Fig. 11). If pure iodine and xenon end members are assumed, an initial $^{129}\text{I}/^{127}\text{I}$ value of 2×10^{-4} is derived (equivalent to some 14 Myr earlier than Shallowater) along with a trapped xenon component with $^{129}\text{Xe}/^{132}\text{Xe} = 0.91$. However, the low inferred value for the trapped component is inconsistent with this interpretation because radioactive decay of ^{129}I can only increase the $^{129}\text{Xe}/^{132}\text{Xe}$ ratio from some initial solar system value such as Q-xenon with $^{129}\text{Xe}/^{132}\text{Xe} = 1.04$ (Busemann et al. 2000) or solar wind with $^{129}\text{Xe}/^{132}\text{Xe} = 1.05$ (Wieler and Baur 1994). Other mechanisms affecting the $^{129}\text{Xe}/^{132}\text{Xe}$ ratio such as mass-dependent fractionation or the incorporation of a chemically fractionated fission xenon component (Shukolyukov et al. 1994) leave characteristic isotopic signatures that are not observed.

The data do not require the end members of the mixing line to be pure iodine and xenon nor is it necessary that all the excess ^{129}Xe present was produced in situ. We can identify a xenon-rich component constrained to lie between points A and B in Fig. 11 and an iodine-rich component similarly constrained to lie between points C and D. Since each component contains iodine, a range of initial iodine values can be derived for each component by projecting a line from a pure xenon component (i.e., Q) through the range defined for each component by the ends of the mixing line to a range

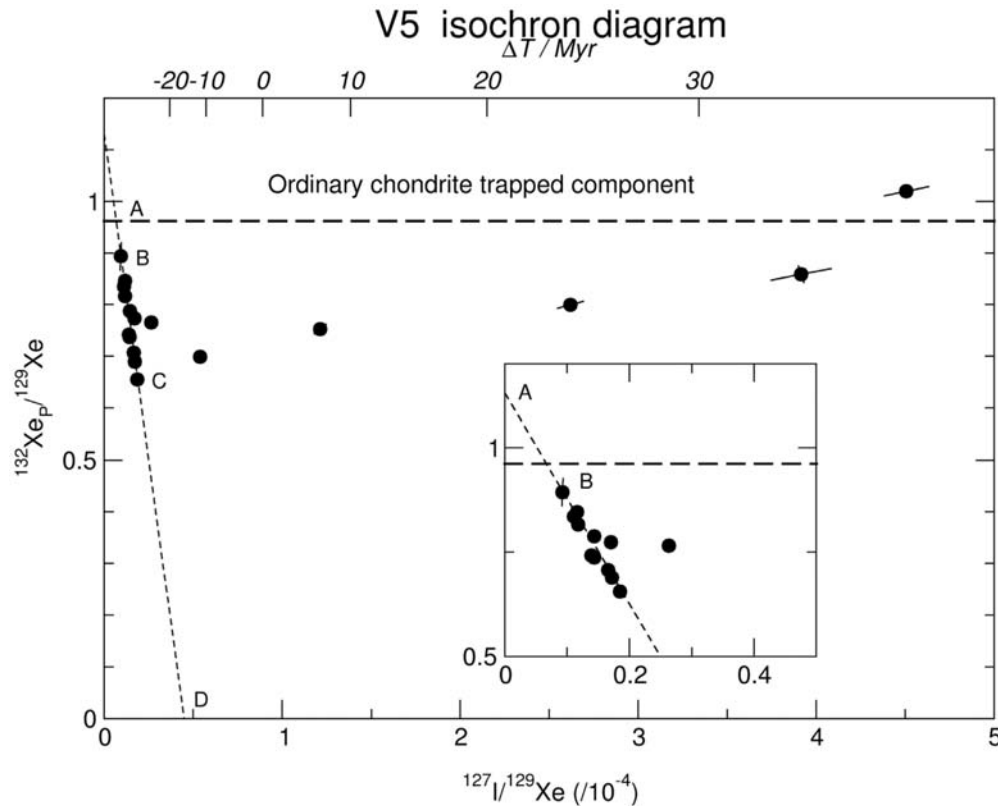


Fig. 11. Three-isotope plot for the Vigarano CAI V5 showing the linear array defined by the high temperature data. The light dashed line is a fit to this data. The intercept on the y-axis implying a trapped xenon component with high $^{132}\text{Xe}_p/^{129}\text{Xe}$ is probably an artefact—see text for discussion. Points A and B define the range of possible compositions for the xenon-rich end member, and C and D define the range for the iodine-rich end member. The heavy dashed line indicates the $^{132}\text{Xe}/^{129}\text{Xe}$ value in Q-xenon or the solar wind. Decay of ^{129}I to ^{129}Xe can only lower this value (the point above this line is due to trapped terrestrial xenon, which is mass fractionated in favor of the heavy isotopes).

of end members assumed to be pure iodine. This procedure leads to constraints on the $^{127}\text{I}/^{129}\text{Xe}$ (equivalently $^{129}\text{Xe}/\text{I}$) ratios of possible end members. The iodine-rich end member calculated in this way must have $^{129}\text{Xe}/\text{I}$ greater than 1.62×10^{-4} which, interpreted chronologically, would correspond to an age more than 8 Myr earlier than Shallowater. In the favored chronology of Gilmour and Saxton (2001) this would lead to an absolute formation age earlier than 4572 Myr, still significantly earlier than the Pb-Pb ages reported for CAIs from CV meteorites (4567.2 ± 0.6 Myr; Amelin et al. 2002; Chen and Wasserburg 1981). Therefore, we are led to seek alternative explanations, unrelated to chronology, to account for this high apparent $^{129}\text{Xe}/\text{I}$ ratio.

The mixing of iodine and xenon to give apparent correlations between ^{129}Xe and ^{128}Xe where there has been demonstrably no in situ decay of ^{129}I is discussed in some detail by Gilmour et al. (2001). The two components that appear to be mixing to form the array in V5 could themselves be mixtures of iodine and xenon. The most iodine-poor point (B in Fig. 11) is consistent with mixing between Q-xenon and iodine with $^{129}\text{I}/^{127}\text{I} \approx 10^{-4}$, such as is observed in several of the Vigarano chondrules (see Fig. 5). Producing the other component (higher I/Xe) from a reservoir with $^{129}\text{I}/^{127}\text{I} = 10^{-4}$ requires a xenon

composition with $^{129}\text{Xe}/^{132}\text{Xe}$ higher than about 1.2. Such components have been observed in, e.g., chondrule V2 from this study as detailed above. This model is capable of explaining the data from the Vigarano CAI in terms of components observed in the other specimens, but no chronological information can be deduced.

Gilmour et al. (2001) described the existence of a mixed component containing iodine and xenon in the Nakhla meteorite and demonstrated that the existence of such components can account for the appearance of iodine and xenon evolving in a closed system. They identified shock incorporation as a mechanism, although in this case, there is no indication that the CAI has experienced a severe shock. Nakhla is, however, only shocked to stage 3, and such a low shock stage cannot be recognized in the opaque probe mount of V5 using conventional techniques (Stöffler et al. 1991). Other mechanisms, notably alteration/deposition of minerals by a brine, may also be capable of producing such mixed components. In addition, alteration of the CAI by a brine that has percolated through a meteorite matrix containing the product of ^{129}I decay provides a natural mechanism for creating a xenon component with an elevated $^{129}\text{Xe}/^{132}\text{Xe}$ ratio (analogous to the common occurrence of “parentless”

^{40}Ar in terrestrial systems). This does not require completely closed system evolution, but a tendency for high $^{129}\text{Xe}/^{132}\text{Xe}$ ratios to be associated with low $^{129}\text{I}/^{127}\text{I}$ ratios is to be expected.

DISCUSSION

Whole Rock Isochrons

The proposed revision of the published whole rock data in the light of re-examination of the I-Xe system in the magnetite standards originally used (Hohenberg et al. 2000; Pravdivtseva et al. 2003a) leads to whole rock ages of 5.4 Myr later than Shallowater for Mokoia and 2.1 Myr later than Shallowater for Vigarano, each with isochrons of good quality (Crabb et al. 1982) and inferred trapped $^{129}\text{Xe}/^{132}\text{Xe}$ ratios that are close to Q-xenon. The I-Xe age-spectra of most of the objects we have examined from these two meteorites reveal significant scatter but are closer to being consistent with the revised ages than with those originally calculated. Thus, we concur with the assessment that the standard in the previous work was unreliable. The contrast between the well-defined whole rock isochrons and the scatter in and among our data for discrete components might be explained if the I-Xe system in the whole rock is dominated by matrix material with a well-defined I-Xe age (such material was not included in our sample suite).

Revising the published whole rock iodine concentrations (Crabb et al. 1982) to be consistent with the new calibration gives values of 51 ppb for Vigarano and 150 ppb for Mokoia, although only 80% and 36%, respectively, of this iodine was found to be correlated with excess ^{129}Xe . These concentrations are somewhat higher than observed for our samples (the mean of all measured Vigarano objects is 21 ppb, the mean of all measured Mokoia objects is 84 ppb, but the CAIs M1 and M2 have concentrations almost as high as the whole rock; see Table 1). The matrix accounts for about 35 vol% of Vigarano and 40 vol% of Mokoia (McSween 1977), while chondrules and CAIs together account for 54 vol% and 51 vol%, so the matrix appears to contain at least half the iodine in Mokoia and a rather greater proportion in Vigarano. Thus, it is reasonable to suppose that the whole rock I-Xe systematics are significantly influenced by the matrix component, even at high release temperatures. Further support for this view arises from the observation that many of our samples show high trapped $^{129}\text{Xe}/^{132}\text{Xe}$ ratios, while the whole rock data is consistent with a trapped $^{129}\text{Xe}/^{132}\text{Xe}$ ratio close to Q-xenon.

In summary, it seems most likely that the appearance of a whole rock isochron from Mokoia and Vigarano is an artefact attributable to most components within them having similar but not identical ages, the high xenon abundance in the whole rock, and the averaging effect of a bulk step heating analysis.

Relative Timing of Alteration

It is clear from mineralogical and petrological evidence that the reduced CV3 lithologies represented by our Vigarano samples have experienced less extensive alteration than the oxidized lithologies such as Mokoia. This is also recorded in lower bulk $^{129}\text{Xe}^*/\text{I}$ ratios of the oxidized group, demonstrating either that their alteration continued after a substantial fraction of ^{129}I had decayed (allowing geochemical separation of daughter xenon from parent iodine) or that early alteration progressively relocated iodine into less retentive sites. These two scenarios are hard to distinguish by examination of the I-Xe system alone, though our studies reveal few samples with straightforward I-Xe systematics, which suggests a complex series of processes. The observation of well-defined I-Xe isochrons for CAIs from Allende (Pravdivtseva et al. 2003b) suggests that the nature or extent of alteration responsible for resetting the I-Xe system was not identical for all CV3_{Ox} meteorites. The complexity of the I-Xe system in the objects considered in this study contrasts with our and other previous studies of suites of chondrules from ordinary and enstatite chondrites, where simple isochrons are the norm rather than the exception. We are led to highlight the role of aqueous processing in the evolution of the carbonaceous chondrites in contrast to the other chondrite groups, where thermal metamorphism has more often played the major role in parent body processing. However, as noted by Swindle et al. (1991), aqueous alteration was the dominant process for the primitive ordinary chondrite Semarkona, which also exhibits complex I-Xe systematics.

Aqueous processing of the carbonaceous chondrites appears to have had a greater effect on the I-Xe system than thermal processing has had in other primitive meteorites. Why should this be so? To answer partially, we must consider the transportation of iodine and xenon by the fluid responsible for aqueous processes. During the heating and cooling cycle of thermal metamorphism, we can envisage a process of resetting of the I-Xe chronometer that ceases as a host mineral cools below the closure temperature. This results in a well-defined resetting event, before which any xenon produced from iodine decay has been lost. In contrast, during aqueous processing, some mineral sites remain unaffected, while others, as a result of chemical reactions, interact with a fluid that may itself contain iodine and $^{129}\text{Xe}^*$ from previous interactions with the host rock. Thus, there is a redistribution during aqueous processing that is not present during thermal metamorphism; this fluid mediated redistribution can also occur on much larger spatial scales than that due to thermal diffusion alone. While the effects of this redistribution can be observed today in “disturbed” I-Xe systematics and trapped components with $^{129}\text{Xe}/^{132}\text{Xe}$ ratios greater than solar (Gilmour et al. 2001), we emphasise the distinction between this and “closed system evolution.” That the CV3_{OxA}

meteorites did not evolve in a closed system is demonstrated by their low bulk $^{129}\text{Xe}^*/\text{I}$ ratios.

I-Xe systematics of chondrules and CAIs from Mokoia and Vigarano suggest that individual objects within the rocks have experienced different histories. In general, the correlation between $^{129}\text{Xe}^*$ (from ^{129}I) and $^{128}\text{Xe}^*$ (from ^{127}I) is rather poor, suggesting an extended history of thermal and chemical processing since the closure to diffusion of the most retentive sites for xenon.

In the case of Mokoia, an I-Xe correlation is observed for both of the measured CAIs, most likely originating from sodalite introduced during aqueous alteration on a parent body. The inferred I-Xe age is 1 Myr later than that of Shallowater. The chondrules from Mokoia, which show little evidence of alteration, do not show a correlation between $^{129}\text{Xe}^*$ and $^{128}\text{Xe}^*$ but appear to have been disturbed at least some 4 Myr later than the CAIs. The whole rock I-Xe “age” of Mokoia of 5.4 Myr later than Shallowater has no simple interpretation.

Some objects from Vigarano have little or no ^{129}Xe excess and have clearly been reset tens of millions of years later than the CAIs in Mokoia. One chondrule (V3) in Vigarano with a well-defined I-Xe age attributable to a metasomatic event at the same time as that observed for the Mokoia CAIs also exists. Anomalies in the I-Xe systematics of CAI V5 make it impossible to interpret the relatively large $^{129}\text{Xe}^*/^{128}\text{Xe}^*$ values chronologically, but this object shows no evidence for late alteration either petrologically or isotopically. These observations suggest that assembly of the chondrules and CAIs in the Vigarano meteorite must have occurred ~100 Myr later than the I-Xe age of Shallowater (sufficient time to allow for the decay of most ^{129}I). Alternatively, some highly localized process must be postulated capable of resetting I-Xe systematics in a chondrule in situ without significantly altering its mineralogy.

On this timescale of ~100 Myr, accretion of planetesimals is expected to be complete, and the resulting planetesimal swarm is well on the way to forming planets. Gravitational effects, and the lack of damping by gas drag allow energetic collisions between objects that are expected to become disrupted and to reassemble under their self-gravity. This would allow the mixing of objects from different positions within a parent body (and from different parent bodies). The late ages of some objects from Vigarano that show no evidence for extensive chemical alteration or prolonged heating imply the existence of an energetic process, probably associated with collisions, capable of resetting I-Xe systematics. The later of the two events recorded in chondrule V2, at 50 Myr later than Shallowater, although interpreted as being due to aqueous alteration, is rather too late to be due to internal heat from radioactive decay in a small body and so may also have been due to energy deposited by a collision.

CONCLUSIONS

CAIs in Mokoia have recorded a clear I-Xe age, probably due to sodalite formation, of approximately 1 Myr later than Shallowater. Scatter about this age may indicate that aqueous processes on the parent body continued for a few million years. The I-Xe systematics of chondrules in both meteorites are, in general, disturbed, although chondrule V3 from Vigarano records an I-Xe age of 0.7 Myr later than Shallowater (also with some scatter), most likely due to aqueous alteration. The contrasting I-Xe systematics in and between chondrules and CAIs suggest that the disturbances predated the consolidation and lithification of the parent bodies. If this is correct, final consolidation occurred at least 4 Myr (Mokoia) and 80 Myr (Vigarano) after the earliest events dated in individual objects from the two meteorites.

I-Xe systematics of the CAI V5 from Vigarano are distinct from all other samples analyzed. A well-defined correlation line appears to correspond to a mixture of trapped xenon with an unacceptably low $^{129}\text{Xe}/^{132}\text{Xe}$ ratio and radiogenic xenon with an improbably high $^{129}\text{I}/^{127}\text{I}$ ratio. An explanation requiring the mixing of trapped xenon and iodine in the same sites, and the presence of parentless ^{129}Xe , means that no chronological information can be obtained from this sample.

I-Xe data from this study also support the assertion by Hohenberg et al. (2000) that the initial iodine value determined by Lewis and Anders (1975) for Murchison magnetite and later adopted as a standard in several I-Xe studies was incorrect. Revised I-Xe ages using the value of Hohenberg et al. (2000) are more easily reconciled with other data.

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Editorial Handling—Dr. Timothy Swindle

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