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An early I-Xe age for CB chondrite chondrule formation, and a re-evaluation of the closure age of Shallowater enstatite

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Abstract–The iodine-xenon system has been analyzed in samples of 7 chondrules from the CB chondrites Gujba and Hammadah al Hamra (HaH) 237. One sample from Gujba defined a high temperature iodine-xenon isochron corresponding to closure 1.87 ± 0.4 Ma before closure of Shallowater enstatite. Motivated by this result, we employ outlier rejection to re-evaluate the Shallowater age, leading to a modified value of 4562.3 ± 0.4 Ma (1σ). In this process, the datum obtained by combining our I-Xe age for Gujba with the literature Pb-Pb age is rejected as an outlier, indicating that in this sample the I-Xe system closed earlier than the accepted Pb-Pb age of chondrules from CB chondrites. The need for a formation environment distinct from that of chondrules from other meteorites is thus reduced.

INTRODUCTION

The CB chondrites are distinguished from other chondrite groups by their high metal content, unusual chondrule textures, strong depletion in moderately volatile lithophile elements, elevated $\delta^{15}N$ (Weisberg et al. 2001), and near absence of fine-grained matrix, though impact melt areas may have been formed from matrix by shock (Meibom et al. 2000). There are two competing models for their formation. In one, it is proposed that they condensed directly from the solar nebula (Weisberg et al. 2001), in the other that they formed from a vapor cloud that resulted from an impact on a pre-existing planetesimal (Krot et al. 2005). Support has been found for the latter theory in the Pb-Pb ages (4562.7 ± 0.25 Myr) for chondrules from the CB chondrites Gujba and Hammadah al Hamra (HaH) 237, even the earliest of which were seen as requiring the nebula to have survived for too long (Krot et al. 2005). This interpretation has been disputed on the grounds that nebulae around young stars can survive for 10 Ma or more (Gounelle et al. 2007). In this work, we present I-Xe data from Gujba and HaH 237 chondrules, which suggest that some CB material predates the Pb-Pb age by at least 1 Ma.

The iodine-xenon system, which was recently reviewed (Gilmour et al. 2006), relies on the decay of ^{129}I to ^{129}Xe, with a half life of 16 Ma. Analyses are similar in principle to those of the Ar-Ar development of the K-Ar system (which was inspired by I-Xe dating). Samples are exposed to a neutron fluence in a reactor. Neutron capture on ^{127}I produces ^{128}I, which β-decays to ^{128}Xe. A correlation is then sought between ^{129}Xe excess (from ^{129}I decay) and ^{128}Xe (produced from iodine in the reactor) across several steps in a step heating experiment. When such a correlation is found, it corresponds to an initial iodine ratio. Relative ages can be inferred on the assumption of isotopic homogeneity of iodine across the relevant region of the solar system. In practice, relative ages are reported as the time intervals that would be required for evolution of the $^{129}I/^{127}I$ ratio between samples and aliquots of Shallowater enstatite included in the same irradiation. Shallowater enstatite has a reproducible initial iodine ratio.

Closure intervals determined from extinct radioisotope chronometers such as the I-Xe system can be converted into absolute ages, provided the absolute age associated with closure of the system in one sample is known. In the case of I-Xe, formation intervals can be converted to absolute ages with reference to the absolute age of the Shallowater standard. At present this is defined from an observed correlation between absolute Pb-Pb ages and I-Xe formation intervals across a range of samples. We return to this calibration below, and suggest it should be modified.

EXPERIMENTAL

Fragments of chondrules from the CB chondrites Gujba and HaH 237 were extracted from polished sections.

Table 1. Summary of data obtained from samples of 7 CB chondrite chondrules.

	^{132}Xe (atoms)		^{128}Xe* (atoms)		^{129}Xe*/^{128}Xe*		^{131}Xe*/^{128}Xe*		^{134}Xe*/^{128}Xe*	
Gujba C1b: 0.449 mg	2.37×10^5	4.7×10^4	2.97×10^6	5.9×10^5	1.925	0.007	0.039	0.0011	0.0195	0.0005
Gujba C2a: 0.444 mg	6.0×10^6	1.2×10^6	3.31×10^5	6.6×10^4	1.05	0.022	2.751	0.057	0.323	0.019
Gujba C2b: 0.649 mg	1.05×10^7	2.1×10^6	5.7×10^5	1.1×10^5	1.877	0.021	1.588	0.04	–	–
HaH 237 C1: 0.5 mg[a]	2.54×10^7	5.1×10^6	1.66×10^7	3.3×10^6	0.118	0.015	2.69	0.22	0.244	0.023
HaH 237 C4: 0.5 mg[a]	2.65×10^7	5.3×10^6	1.26×10^7	2.5×10^6	0.089	0.021	1.83	0.11	0.297	0.025
Gujba C2a: 0.5 mg[a]	5.5×10^7	1.1×10^7	3.07×10^7	6.1×10^6	–	–	–	–	–	–
Gujba C1: 1.2 mg	1.89×10^6	3.8×10^5	6.7×10^4	1.3×10^4	0.35	0.13	0.62	0.27	1.52	0.15

^{128}Xe* represents ^{128}Xe excess over a Xe-Q composition, and so on. ^{128}Xe* is derived from iodine, ^{131}Xe* from barium or tellurium, and ^{134}Xe* from uranium. [a]Precise masses of these three samples are unavailable, but their masses were similar to the preceding three Gujba samples.

Chondrules in our Gujba sample (which was sourced from E. Twelker) were identified as pale rounded silicates with a peak diameter close to 3 mm embedded in a dark matrix. Chondrules in HaH 237 (provided by the Natural History Museum, London, sample BM2000, M9) were similar, though the mean chondrule diameter in our sample was slightly smaller. Gujba sample C1 was included in irradiation MN17, samples C1b, C2a, and C2b were part of irradiation MN18, while sample C2c was part of irradiation MN19, which also included two samples of HaH 237 (C1 and C4). ^{128}Xe*/^{129}Xe* ratios for Shallowater (where * denotes amounts of the isotope produced from iodine) were 0.531 ± 0.007, 0.514 ± 0.009, and 0.540 ± 0.023 for MN17, MN18 and MN19, respectively. This allows the conversion efficiency of ^{127}I to ^{128}Xe* to be calculated with reference to the ^{129}I/^{127}I ratio of Shallowater enstatite. For this conversion, we adopt a ^{129}I/^{127}I ratio of 1.072×10^{-4} for Shallowater enstatite (Nichols et al. 1994); since we are interested only in relative initial iodine ratios, uncertainty in this quantity is neglected.

After irradiation, samples were loaded into the laser port of the RELAX (Refrigerator Enhanced Laser Analyser for Xenon) mass spectrometer (Gilmour et al. 1994). The analyses reported here were performed before the instrument upgrades recently reported by Crowther et al. (2008), though the data reduction procedure described therein was adopted to generate isotope ratios and absolute amounts of gas. Gas was extracted by heating the samples for one minute at each of a series of sequentially increasing laser powers. Released gas was gettered for one minute to remove active gases, then admitted to the mass spectrometer whereupon data acquisition began. Data were acquired in 30 ten-second segments over a period of five minutes. Isotope ratios and the height of a normalizing isotope were extrapolated to the time of gas inlet from corresponding quantities determined for each of these segments. Absolute gas quantities were calculated and a mass discrimination correction made with reference to interspersed air calibrations. Procedural blank was monitored but made negligible contribution to the isotopic data discussed here. All errors discussed in this paper are 1 standard deviation. Data are summarized in Table 1, and complete data tables are available in accompanying supplementary information.

RESULTS AND DISCUSSION

Data show clear evidence of a contribution from fission of an isotope of uranium or plutonium (Fig. 1). Possible candidates include a mixture of spontaneous fission xenon from ^{244}Pu and ^{238}U parents, or neutron-induced fission of ^{235}U, with a composition modified by neutron capture on fissiogenic ^{135}Xe during the irradiation (Ozima and Podosek 1983). Other samples (of lodranites; which will be reported elsewhere) included in these same irradiations are consistent with the fission correlation line derived from the CB chondrite data and have ^{134}Xe/^{132}Xe ratios greater than those associated with either of the spontaneous fission parents (but less than that of neutron-induced fission of ^{235}U). We conclude that ^{132}Xe in these samples is predominantly a mixture of trapped xenon, for which we adopt the composition of Q-Xe (^{134}Xe/^{132}Xe = 0.3780 ± 0.0011 (Busemann et al. 2000), and xenon from neutron-induced fission of ^{235}U with ^{134}Xe/^{132}Xe = 1.840 ± 0.076 (Ozima and Podosek 1983); ^{132}Xe associated with the trapped component was calculated on this basis, and excesses of ^{128}Xe, ^{129}Xe and ^{134}Xe discussed below were calculated over the Q-Xe composition associated with the trapped component. To allow comparison of data across three irradiations, ^{128}Xe*/^{129}Xe was converted to I/^{129}Xe based on the I-Xe correlation lines from the relevant Shallowater samples.

Data from all seven samples are presented in Fig. 2. Sample C1b yields an excellent isochron from 5 consecutive high temperature releases (MSWD = 0.55 where deviates exclude the systematic error from the conversion of ^{128}Xe* to iodine). No other sample yielded an isochron. The C1b isochron corresponds to a (nominal) initial iodine ratio of $(1.16 \pm 0.02) \times 10^{-4}$ or, equivalently, closure 1.87 ± 0.4 Ma before closure of Shallowater enstatite. Adopting the absolute date of Shallowater closure proposed by Gilmour et al. (2006) (4563.3 ± 0.4) this corresponds to closure at 4565.2 ± 0.6 Myr. A few individual releases from other samples correspond to earlier model ages when considered as a mixture of iodogenic xenon and Q-Xe, but shock disturbance of the system or differential mobilization of iodine and xenon can explain such observations, and chronological significance cannot be concluded in the absence of an isochron. The only chronological constraint arising from these data is that associated with the isochron from Gujba chondrule C1b.

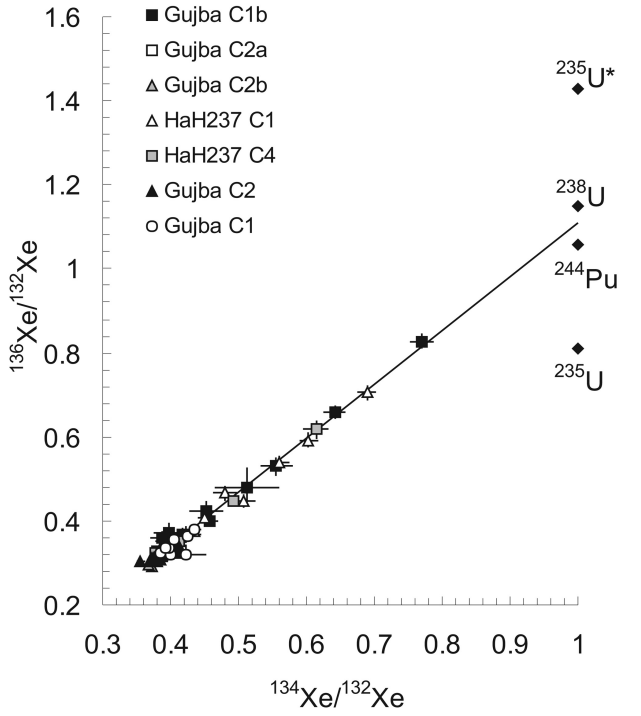

Fig. 1. Several samples indicate a contribution from fission to the heavy xenon isotopes. The compositions of notional components that are a mixtures of Q-Xe and fission xenon with $^{134}Xe/^{132}Xe = 1$ are marked with the corresponding parent. The observed fission composition might be explained by a mixture of xenon from spontaneous fission of ^{238}U and ^{244}Pu. However, in high flux reactions, the composition of xenon produced by neutron-induced fission of ^{235}U is modified by production of ^{136}Xe from neutron capture on $^{135}Xe^{*}$—complete conversion of the 135 isobar to 136 is represented by the composition labelled $^{235}U^{*}$. Other samples in the same irradiations lay on the same correlation line but with $^{134}Xe/^{132}Xe$ ratios higher than consistent with either spontaneous fission endmember, identifying the parent as ^{235}U.

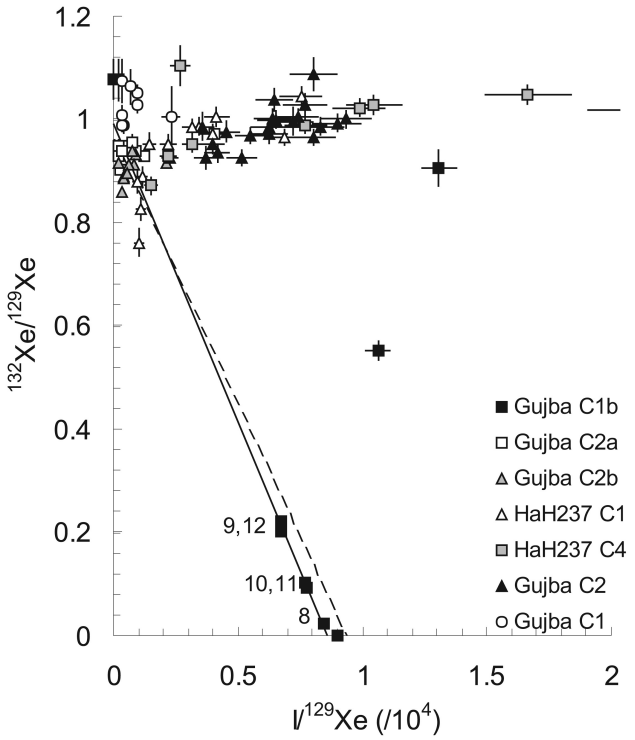

Fig. 2. Accumulated I-Xe data for the seven chondrule fragments analyzed. (Only) Gujba chondrule C1b exhibits an isochron (solid line), which was formed by the 5 highest temperature data points (labelled with release numbers). The isochron is distinct from that of Shallowater enstatite (broken line). The corresponding closure interval is 1.87 ± 0.4 Myr, corresponding to an initial $^{129}I/^{127}I$ ratio of $1.16 \pm 0.02 \times 10^{-4}$. The remaining data show evidence of uncorrelated iodine mixing with a trapped component with a composition ($^{132}Xe/^{129}Xe = 0.95 \pm 0.03$) close to that of Xe-Q. Some data from other chondrule fragments exhibit excesses of ^{129}Xe, but in the absence of isochronous behavior chronological interpretation is unjustified.

Other chronological constraints on the CB chondrites arise from the Al-Mg and U-Pb/Pb-Pb systems. Gounelle et al. (2007) reported that the initial $^{26}Al/^{27}Al$ ratio was less than 8×10^{-6}, which corresponds to an interval of >2.1 Myr after the canonical ratio associated with CAI formation. Adopting a Pb-Pb age of CAIs of 4567.2 ± 0.6 (Amelin et al. 2002) this corresponds to an absolute age of $<4565.1 \pm 0.6$ Myr, borderline consistent with our I-Xe age. Krot et al. (2005) reported an earliest Pb-Pb age of 4562.7 ± 0.25 Myr (1σ) for Gujba chondrules, which they identified with chondrule formation. The earliest absolute age we derive from I-Xe data is earlier than this, though both the I-Xe system and Pb-Pb systems suggest ongoing processing after chondrule formation. This prompts us to consider whether the absolute age (and associated error) of the Shallowater standard needs updating, and whether any modification can account for the discrepancy between the Pb-Pb and I-Xe systems in Gujba.

Typically, deriving absolute ages from a short-lived chronometer involves defining one sample as an "anchor" in which it is proposed that the short-lived system dates the same event as the Pb-Pb chronometer. The ages of other samples in the short-lived system are then converted to absolute ages with reference to this anchor. We believe that a better approach is to plot absolute Pb-Pb ages against corresponding intervals in the short-lived system, the presence of a correlation is a test of the hypothesis that the two systems evolved coherently and the associated relationship allows the absolute age of the standard and its uncertainty to be calculated.

For instance, in calculating the current best estimate of the age of Shallowater enstatite, Gilmour et al. (2006) sought a correlation between formation intervals in the I-Xe system and absolute ages in the long-lived system across a range of samples that represented all available possible calibration points. They found that, while a correlation was present, the scatter of points around the line was greater than would be expected based on the precision of the measurements. This suggests the presence of outliers—some

Table 2. I-Xe interval, Pb-Pb ages and corresponding ages of the Shallowater standard.

	I-Xe interval		Pb-Pb age		Shallowater age	
Earliest chondrules (Amelin and Krot 2007; Swindle et al. 1991a, 1991b)	4.3	0.6	4566.6	1.0	4562.4	1.2
Ste. Marguerite whole rock (Brazzle et al. 1999; Göpel et al. 1994)	0.7	0.4	4566.7	0.8	4566.0	0.9
Richardton chondrule (Gilmour et al. 2006)	–0.1	0.1	4563.2	0.8	4563.3	0.8
Ste. Marguerite phosphate (Brazzle et al. 1999; Göpel et al. 1994)	0.7	0.4	4562.7	0.3	4562.0	0.5
Gujba (this work, Krot et al. 2005)	1.9	0.4	4562.7	0.3	4560.8	0.5
Richardton pyroxene (Amelin 2001; Pravdivtseva et al. 1998)	1.1	2.0	4562.2	0.6	4561.1	2.1
Richardton chondrule (Gilmour et al. 2006)	–4.1	0.6	4559.0	1.4	4563.1	1.5
Acapulco phosphate (Brazzle et al. 1999; Göpel et al. 1994; Nichols et al. 1994)	–8.8	0.2	4557.0	1.0	4565.8	1.0
Acapulco feldspar (Brazzle et al. 1999; Göpel et al. 1994)	–3.8	1.5	4557.0	1.0	4560.8	1.8
Kernouve phosphate (Brazzle et al. 1999; Göpel et al. 1994)	–43.0	6.0	4522.5	0.7	4565.5	6.0

I-Xe data from the literature and this work and corresponding literature Pb-Pb ages are combined to produce a Pb-Pb age equivalent to closure of the I-Xe system in Shallowater enstatite.

members of the data set are not consistent with coherent evolution of the two chronometers. This is inevitably the case since more than one candidate I-Xe age for Acapulco is compared to the Pb-Pb phosphate age and more than one candidate Pb-Pb age is compared to the Ste Marguerite I-Xe age—this reflects uncertainty as to the closure temperatures of the chronometers involved in different mineral systems. They recommended a Shallowater age and precision based on the effects of excluding various points from the correlation based on various hypotheses as to the cause of the excess scatter.

We now attempt to revise this estimate by excluding outliers from the correlation statistically. To this end we will employ a modified data set (Table 2) including only those samples for which I-Xe and Pb-Pb ages are available in the literature (and this work) that at least plausibly record the same event. Our base data set includes the Gujba I-Xe interval of this work combined with the Pb-Pb age of Krot et al. (2005) and the data set of Gilmour et al. (2006) where we adopt the 4566.7 ± 1.0 Pb-Pb age of Amelin et al. (2007) for the oldest chondrules. Since the Mn-Cr to I-Xe conversion is on a less certain footing (Busfield et al. 2008), we exclude data derived indirectly via Mn-Cr that were included in the previous work.

In seeking to distinguish outliers that disproportionately affect the correlation, we note that each sample with an I-Xe and Pb-Pb age (each with analytical uncertainties) corresponds to a hypothetical age for the Shallowater standard and associated error. We treat this as a set of observations and evaluate the likelihood that models where the U-Pb and I-Xe systems evolve coherently can account for the data, the models being distinguished from one another by differing absolute ages for the Shallowater standard. The process of choosing a model with maximum likelihood is equivalent to that of fitting a line of gradient one to the data in Fig. 3 by minimizing χ^2.

To proceed, we exclude each datum in turn from our base data set. We then determine the maximum likelihood model for this reduced data set. The maximum likelihood of each reduced data set is plotted in Fig. 4 as a multiple of the maximum likelihood of the base data set. In Fig. 4 it is apparent that removal of any one of three samples has a much more dramatic effect on our ability to account for the data with a common model than removal of one of the other seven. These samples are Ste. Marguerite whole rock, Gujba and Acapulco apatite. Exclusion of all of these three points from the data set leads to MSWD = 0.7 when a Williamson method is used to fit a correlation line to the remaining data (Fig. 5) and an estimate for the absolute age of the Shallowater standard of 4562.3 ± 0.4 Ma, 1 million years more recent than that previously proposed. We also conclude that the Ste. Marguerite whole rock Pb-Pb age predates its phosphate I-Xe age; the Acapulco phosphate I-Xe age was set later than its Pb-Pb age, which is in fact more consistent with the I-Xe age of the feldspar; the Gujba chondrule Pb-Pb age postdates the I-Xe age of the chondrule studied in this work.

The above discussion indicates that associating the Pb-Pb and I-Xe ages of our Gujba chondrule leads to a relationship that is not consistent with the majority of the remaining samples. Adopting the proposed new calibration, we determine that the absolute I-Xe closure age of the Gujba chondrule is 4564.2 ± 0.6. This is clearly consistent with the limit imposed from the Al-Mg system, but still significantly older than the proposed Pb-Pb age of chondrule formation by approximately 1.5 Myr. We consider it unphysical that the

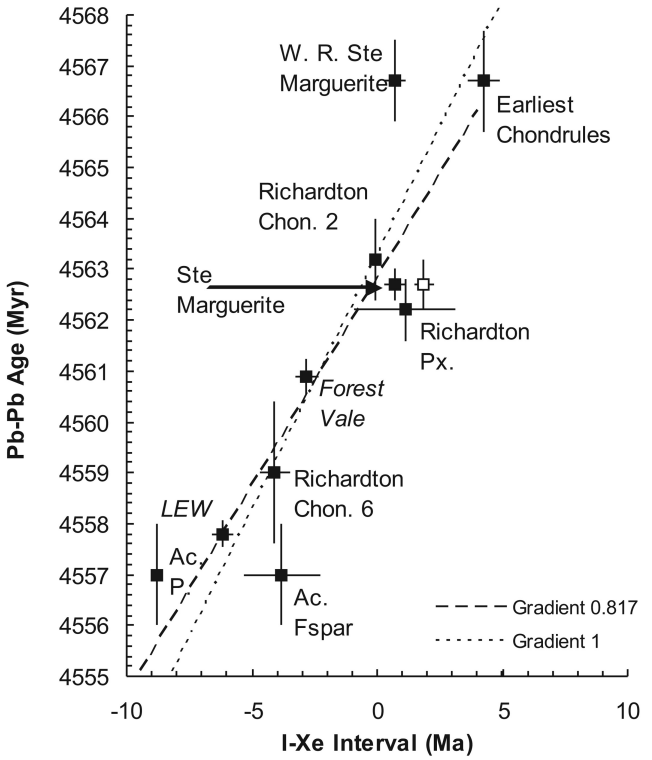

Fig. 3. Comparison of Pb-Pb and I-Xe ages in a range of material from the early solar system. The lines correspond to the results of fits to literature data (solid symbols, Table 2—where references to data sources are to be found) with gradient optimized (best fit gradient = 0.817) and gradient defined to be 1 (as required for concordant evolution). The datum obtained by combining the I-Xe age of Gujba C1b (this work) with the absolute age of Krot et al. (2005) is shown with an open symbol and is inconsistent with the line defined by the other data. Abbreviations: W. R. = whole rock; Chon. = chondrule; Px = pyroxene; LEW = Lewis Cliff 86010; Ac. P. = Acapulco phosphate; Ac. Fspar = Acapulco feldspar.

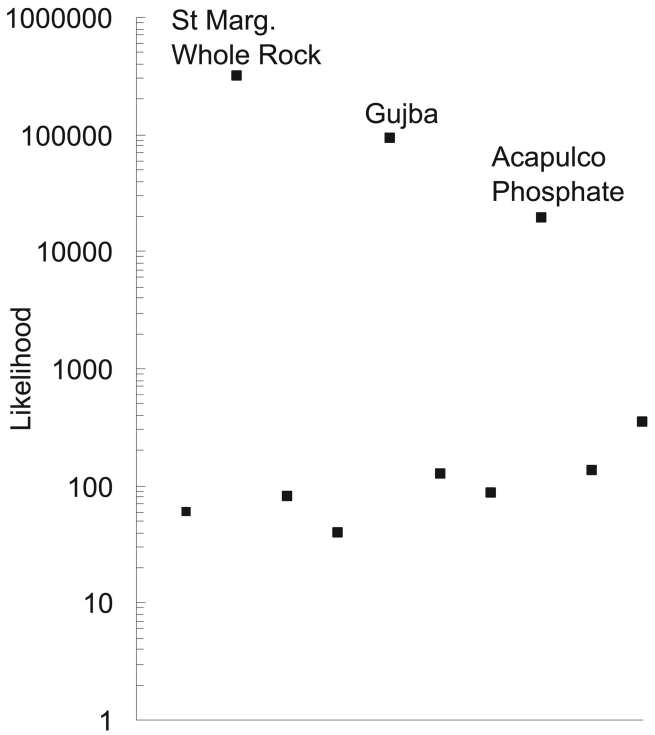

Fig. 4. The data set of Table 2 can be used to determine the maximum likelihood model of coherent evolution of the I-Xe and Pb-Pb systems. To examine which points have a disproportionate effect, we exclude each datum in turn and fit a model to the remaining data by maximizing the likelihood. The factor by which this maximum likelihood exceeds that of a model fit to the base data set that includes all data is plotted here—from left to right samples are in the same sequence as listed in Table 2. The relative increase in maximum likelihood for three samples dramatically exceeds that for the rest: Ste. Marguerite whole rock, Gujba, Acapulco phosphate. Exclusion of all 3 samples leads to the correlation line of Fig. 5.

I-Xe system records an event before chondrule formation, since this would require that a grain or grains within the chondrule were not degassed by a melting event at temperatures comparable to those employed to extract gas in our laboratory. We also note that our absolute I-Xe age is in excellent agreement with the Pb-Pb age reported for HaH 237 by the same authors. We therefore conclude that the Pb-Pb system in the three chondrules reported by Krot et al. (2005) records an event after chondrule formation. It is interesting that the release of uranium-derived $^{134}Xe^{*}$ (like that of iodine-derived $^{128}Xe^{*}$) from our sample was bimodal suggesting the presence of two uranium carrier phases, one of which has iodine with no detectable ^{129}Xe, while the I-Xe isochron selects only the high release temperature site (Fig. 6). We note that the other chondrules studied by Krot et al. yielded younger Pb-Pb ages, suggesting that processes capable of disturbing/resetting the U-Pb system were ongoing on the CB chondrite parent body after chondrule formation and that the Pb-Pb system, like the I-Xe system, reflects ongoing processes over a significant interval. In such circumstances, the oldest isochron age represents a most recent limit on chondrule formation.

Although Gounelle et al. (2007) have already argued that the nebula might have persisted beyond the time of the Pb-Pb age, revising the formation age of CB chondrite chondrules to an earlier date than is suggested by the Pb-Pb age lessens the need for a distinct formation mechanism involving collisions. In addition, evidence that the I-Xe and Pb-Pb systems are discordant in CB chondrites, and that both show evidence of processing after chondrule formation, suggests that they may not be ideal candidates for future attempts to relate other chronological systems based on extinct radioisotopes to one another and to Pb-Pb.

It is notable that the revision of the absolute age adopted for Shallowater based on the outlier rejection approach exceeds the range of uncertainties derived in both the previous and present analyses. This reflects the fact that such errors reflect uncertainty in a parameter, given that the assumptions employed during data reduction are valid. The disadvantage of a process such as conventional line fitting that maximizes the likelihood of a model is that it is sensitive

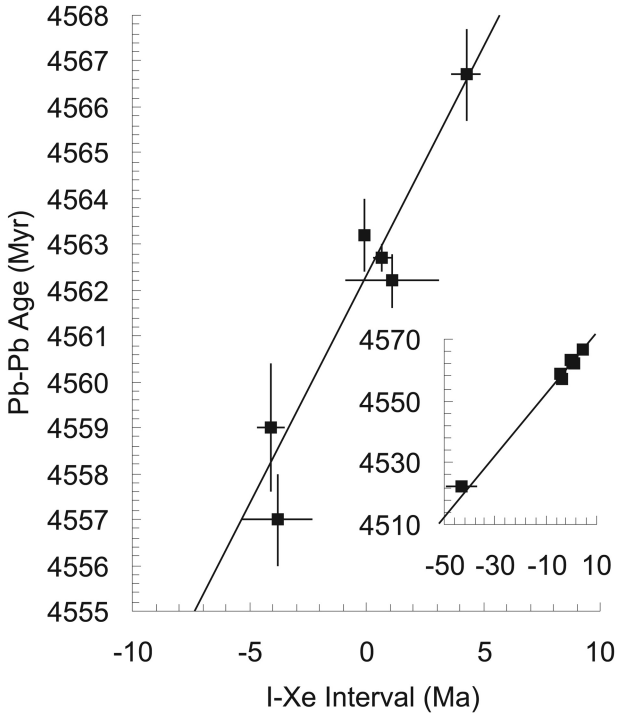

Fig. 5. Correlation line fitted through the reduced data set as defined in Fig. 4. If the gradient is defined to be 1 (as expected for coherent evolution of U-Pb and I-Xe), the closure age of Shallowater enstatite is 4562.3 ± 0.4 with MSWD (mean square weighted deviate) = 0.7. Allowing the gradient to vary in the fit yields a best fit gradient of 0.96 ± 0.11 and an identical intercept and MSWD. (The inset graph shows an expanded scale allowing display of the Kernouve datum. Samples can be identified with reference to Fig. 3, where they are labelled.)

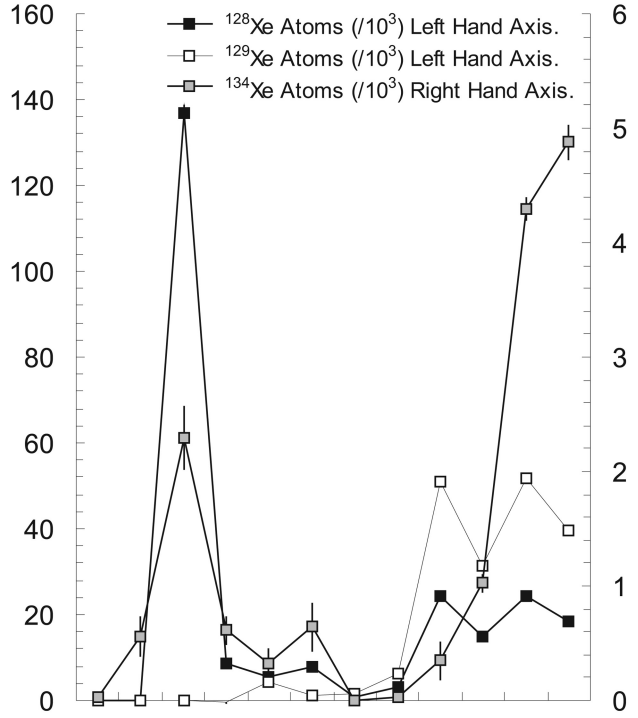

Fig. 6. Release patterns from chondrule C1b of isotopes produced in the reactor from ^{127}I (^{128}Xe) and ^{235}U (^{134}Xe) and ^{129}Xe (produced by ^{129}I decay in the early solar system). The power of the continuous wave laser with which the samples were heated, and thus the temperature, increases from left to right, but temperature data are not available. Both iodine and uranium are present in a low temperature site which has a more recent I-Xe model age than the site responsible for the high temperature isochron, uranium also appears to be present in a site with a higher xenon release temperature than that responsible for the I-Xe isochron.

to the inclusion of data that are mistakenly identified as part of the distribution. The disadvantage of outlier rejection such as that employed here and similar to that adopted in some Pb-Pb studies is that it assumes that some data are not representative of the correlation when in fact it is only unlikely that they are part of the correlation. Our quoted precision on the Shallowater calibration age is predicated on the assumption that some data points ought to be excluded—the small but finite chance that they are part of the distribution is excluded so the probability that the calibration lies within the quoted interval is overestimated. As always, the assumptions used in deriving a parameter and its quoted uncertainty should not be forgotten. Nonetheless, the balance of evidence currently indicates that the Pb-Pb age of chondrules from CB chondrites is not representative of chondrule formation.

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Editorial Handling—Dr. Timothy Swindle

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