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Meteorites and the Early Solar System II

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Timescales and Settings for Alteration of Chondritic Meteorites

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Most groups of chondritic meteorites experienced diverse styles of secondary alteration to various degrees that resulted in formation of hydrous and anhydrous minerals (e.g., phyllosilicates, magnetite, carbonates, ferrous olivine, hedenbergite, wollastonite, grossular, andradite, nepheline, sodalite, Fe,Ni-carbides, pentlandite, pyrrhotite, Ni-rich metal). Mineralogical, petrographic, and isotopic observations suggest that the alteration occurred in the presence of aqueous solutions under variable conditions (temperature, water/rock ratio, redox conditions, and fluid compositions) in an asteroidal setting, and, in many cases, was multistage. Although some alteration predated agglomeration of the final chondrite asteroidal bodies (i.e. was pre-accretionary), it seems highly unlikely that the alteration occurred in the solar nebula, nor in planetesimals of earlier generations. Short-lived isotope chronologies (^{26}Al - ^{26}Mg , ^{53}Mn - ^{53}Cr , ^{129}I - ^{129}Xe) of the secondary minerals indicate that the alteration started within 1-2 Ma after formation of the Ca,Al-rich inclusions and lasted up to 15 Ma. These observations suggest that chondrite parent bodies must have accreted within the first 1-2 Ma after collapse of the protosolar molecular cloud and provide strong evidence for an early onset of aqueous activity on these bodies.

1. INTRODUCTION

In this chapter, we review the mineralogy, petrology, and time-scales for secondary alteration of type 1-3 carbonaceous (CI, CM, CR, CV, and ungrouped carbonaceous chondrite MAC88107), enstatite and ordinary chondrites that resulted in the formation of hydrous and anhydrous minerals (e.g., phyllosilicates, carbonates, magnetite, Ni-bearing sulfides, fayalite, ferrous olivine, andradite, hedenbergite, wollastonite, grossular, nepheline, sodalite). Although thermal and shock metamorphism are also among the secondary processes which affected most chondritic meteorites and resulted in some mineralogical modifications, the ages of these processes are not discussed here.

Chondrites consist of four major components: chondrules, Fe,Ni-metal grains and/or metal-troilite aggregates, refractory inclusions [Ca,Al-rich inclusions (CAIs) and amoeboid olivine aggregates (AOAs)], and fine-grained matrix material. The only exception is CI chondrites which lack chondrules, refractory inclusions, and Fe,Ni-metal grains. In addition, some chondrites contain foreign lithic clasts. It is generally believed that the refractory inclusions, chondrules, and Fe,Ni-metal formed in the solar nebula by high temperature processes that included evaporation and condensation. Many CAIs and most chondrules and Fe,Ni-metal were subsequently melted during multiple brief heating episodes. The refractory inclusions are considered to be the oldest solids formed in the solar nebula 4567.2 ± 0.6 Ma ago (*Amelin et al.*, 2002). Chondrule formation appears to have started less than 1 Myr after CAIs and lasted for at least 4 Myr (*Amelin et al.*, 2004; *Bizzarro et al.*, 2004). Chondrules and matrices in a primitive chondrite are chemically complementary (*Bland, pers. comm.*, 2004), suggesting that most of the matrix materials could have been thermally processed during chondrule formation (*Scott and Krot*, 2004).

Most chondrite groups show evidence for relatively low temperature alteration that affected all their chondritic components (*Brearley and Jones, 1998*). The nature of this alteration remains controversial and has been attributed to nebular (or pre-accretionary) and/or asteroidal processing (e.g., *Brearley, 2003*). Timing of the alteration using short-lived chronology such as ^{26}Al - ^{26}Mg , ^{53}Mn - ^{53}Cr , and ^{129}I - ^{129}Xe , can potentially resolve this controversy and constrain ages of chondrule formation and time of accretion of the chondrite parent asteroids. We note that because the life time of the solar nebula is poorly constrained (*Podosek and Cassen, 1994*), dating of secondary alteration alone typically cannot distinguish between nebular and asteroidal settings of alteration, which should be based on mineralogical and isotopic (e.g., oxygen) observations, thermodynamic analysis, and petrologic experiments. At the same time, the prolonged duration of alteration and similar ages of alteration to other asteroidal processes, such as thermal metamorphism and igneous differentiation, favor asteroidal settings of alteration. Since the environment of alteration (nebular vs. asteroidal) remains controversial, in each section we briefly summarize the mineralogical, petrologic and isotopic (oxygen isotopic compositions) arguments supporting nebular or asteroidal settings for the alteration of a chondrite group (see also *Brearley, 2003, 2005*).

2. SHORT-LIVED ISOTOPE CHRONOLOGY OF SECONDARY ALTERATION OF CHONDRITIC METEORITES

2.1. ^{26}Al - ^{26}Mg Ages

^{26}Al is a short-lived radionuclide that β -decays to ^{26}Mg with a half-life of ~ 0.73 Ma. Excess ^{26}Mg ($^{26}\text{Mg}^*$) can be detected by secondary ionization mass spectrometry (SIMS) or by other mass spectrometric techniques in bulk samples or mineral fractions [e.g., thermal

ionization mass spectrometry (TIMS) and inductively coupled plasma mass spectrometry (ICP-MS)]. If $^{26}\text{Mg}^*$ is derived from *in situ* decay of ^{26}Al , then the data points plotted as $\delta^{26}\text{Mg}$ [permil (‰) deviation from the terrestrial $^{26}\text{Mg}/^{24}\text{Mg}$ ratio of 0.13932] against the $^{27}\text{Al}/^{24}\text{Mg}$ ratio will define a straight line (Al-Mg isochron) with the slope proportional to $^{26}\text{Al}/^{27}\text{Al}$ at the time of Al-Mg isotope system closure. Based on the measured abundances of $^{26}\text{Mg}^*$ in numerous CAIs, the solar system initial $^{26}\text{Al}/^{27}\text{Al}$ ratio, called "canonical", is estimated to be $\sim 5 \times 10^{-5}$ (e.g., *MacPherson et al.*, 1995; *Bizzarro et al.*, 2004). The difference in the initial $^{26}\text{Al}/^{27}\text{Al}$ ratios between the unknown sample and the canonical $^{26}\text{Al}/^{27}\text{Al}$ ratio in CAIs corresponds to their relative formation age:

$$(1) \quad \Delta t_{\text{sample-CAI}} (\text{Ma}) = 1/\lambda \times \ln[(^{26}\text{Al}/^{27}\text{Al})_{\text{CAI}}/(^{26}\text{Al}/^{27}\text{Al})_{\text{sample}}],$$

where $\lambda = \ln 2/0.73$ is the ^{26}Al decay constant; negative/positive values correspond to older/younger ages than CAIs with a canonical $^{26}\text{Al}/^{27}\text{Al}$ ratio.

2.2. ^{53}Mn - ^{53}Cr Ages

^{53}Mn is a short-lived radionuclide that β -decays to ^{53}Cr with a half-life of ~ 3.7 Ma (*Lugmair and Shukolyukov*, 1998). This half-life and the fact that Mn and Cr are reasonably abundant elements that experienced extensive fractionation during aqueous alteration, make the ^{53}Mn - ^{53}Cr chronometer very useful for dating aqueous activity on chondrite parent asteroids (e.g., formation of carbonates and fayalite).

The excess of ^{53}Cr ($^{53}\text{Cr}^*$) relative to the terrestrial $^{53}\text{Cr}/^{52}\text{Cr}$ ratio of 0.113458 (*Papanastassiou*, 1986) can be detected by SIMS in individual minerals having high (>100) Mn/Cr ratios, which can yield a high concentration of radiogenic ^{53}Cr , with minimal interference from non-radiogenic Cr. If $^{53}\text{Cr}^*$ is derived from *in situ* decay of ^{53}Mn , then the data points

plotted as $\delta^{53}\text{Cr}$ (% deviation from the terrestrial $^{53}\text{Cr}/^{52}\text{Cr}$ ratio) against $^{55}\text{Mn}/^{52}\text{Cr}$ ratio will define a straight line (Mn-Cr isochron) with the slope proportional to $^{53}\text{Mn}/^{55}\text{Mn}$ ratio at the time of the isotope closure of Mn-Cr system. The relative ages of two samples, 1 and 2, are then calculated from their $^{53}\text{Mn}/^{55}\text{Mn}$ ratios:

$$(2) \quad \Delta t_{1-2} (\text{Ma}) = 1/\lambda \times \ln[(^{53}\text{Mn}/^{55}\text{Mn})_2 / (^{53}\text{Mn}/^{55}\text{Mn})_1],$$

where $\lambda = \ln 2/3.7$ is the ^{53}Mn decay constant. Due to the uncertainty in the solar system initial abundance of ^{53}Mn [estimates range from 0.84×10^{-5} (*Lugmair and Shukolyukov, 1998*) to 1.4×10^{-5} (*Lugmair and Shukolyukov, 2001*) to $(2.8 \pm 0.3) \times 10^{-5}$ (*Nyquist et al., 2001*) to 4.4×10^{-5} (*Birck and Allègre, 1988; Birck et al, 1999*)], the Mn-Cr ages discussed below are given relative to the $(^{53}\text{Mn}/^{55}\text{Mn})_0$ ratio of $(1.25 \pm 0.07) \times 10^{-6}$ for the angrite Lewis Cliff (LEW) 86010 (Δt_{LEW}) that has the absolute age determined by Pb/Pb of 4557.8 ± 0.5 Ma (*Lugmair and Shukolyukov, 1998*).

2.3. ^{129}I - ^{129}Xe Ages

The incorporation of live ^{129}I into solid matter in the early solar system and subsequent β -decay resulted in production of its stable ^{129}Xe daughter ($^{129*}\text{Xe}$) at iodine-bearing sites. Isotopic closure, achieved when I and Xe migration ceased, preserved a parent-daughter record that is observable today in whole-rock samples or mineral separates in many meteorites. If no Xe losses have occurred, the ratio of radiogenic $^{129*}\text{Xe}$ to stable ^{127}I equals the value for the initial iodine ($^{129}\text{I}/^{127}\text{I}$) at the time of isotopic closure. Due to the 15.7 Ma half-life of ^{129}I , $^{129}\text{I}/^{127}\text{I}$ evolved rapidly in the early solar system. Differences in this initial iodine among meteoritic samples form the basis of I-Xe dating (*Reynolds, 1960*). The analytical technique of I-Xe dating involves neutron irradiation in a reactor, which converts a fraction of ^{127}I to $^{128*}\text{Xe}$ [$^{127}\text{I} (n, \gamma\beta) \rightarrow ^{128*}\text{Xe}$]. Correlated quantities of two iodine derived Xe isotopes ($^{129*}\text{Xe}$ and $^{128*}\text{Xe}$) released in stepwise

pyrolysis and measured by ion counting mass spectrometry (Hohenberg, 1980). The simplicity of this technique is enhanced by including in the irradiation a meteorite standard of known age (Nichols *et al.*, 1994) with the relative I-Xe age then given by the relative slopes of the isochrons. Typically, the ratio of ^{129}Xe to some Xe isotope not produced in the irradiation, such as ^{130}Xe or ^{132}Xe , is plotted against the ratio of ^{128}Xe to that same isotope. The choice of ^{130}Xe or ^{132}Xe normalization is usually determined by the relative correction (if any) for spallation or fission effects, respectively, at these isotopes. If the $^{128*}\text{Xe}$ and $^{129*}\text{Xe}$ are both derived from iodine of uniform isotopic composition, then the data points will define a straight line (I-Xe isochron), with the slope proportional to the $^{129}\text{I}/^{127}\text{I}$ ratio at the last time Xe isotopes were in equilibrium (Swindle and Podosek, 1988). The I-Xe isochron is thus a two-component mixture of trapped and I-derived Xe. The trapped Xe component is confined to lie at the lower end of this isochron and typically of “planetary” composition (Lavielle and Marti, 1992). Therefore, I-Xe ages are calculated directly from the differences in isochron slopes $(^{129*}\text{Xe}/^{128*}\text{Xe})_{\text{sample}}$ with that of the standard $(^{129*}\text{Xe}/^{128*}\text{Xe})_{\text{standard}}$ (Shallowater aubrite or Bjurböle L4 ordinary chondrite; Bjurböle predates Shallowater by 460,000 yrs, Brazzle *et al.*, 1999):

$$(3) \quad \Delta t_{\text{sample-Shallowater}} (\text{Ma}) = 1/\lambda \times \ln[(^{129}\text{I}/^{127}\text{I})_{\text{Shallowater}}/(^{129}\text{I}/^{127}\text{I})_{\text{sample}}],$$

where $\lambda = \ln 2/15.7$ is the ^{129}I decay constant; negative/positive values correspond to older/younger ages than Shallowater. Based on the comparison of I-Xe and Mn-Cr systems with the absolute Pb-Pb chronometer for samples analysed by multiple systems, Gilmour *et al.* (work in progress) concluded that the I-Xe system closed in Shallowater aubrite 5.7 ± 1.1 Ma earlier than the Mn-Cr system closed in LEW86010 angrite, at 4563.5 ± 1.0 Ma before the present. Use of St. Severin as internal standard during the early measurements (e.g., Zaikowski, 1980), which was later shown to be inhomogeneous (Hohenberg *et al.*, 1981), makes these measurements difficult

to compare with recent results, although the relative ages should be meaningful (*Pravdivtseva et al.*, 2003a).

Because iodine is a mobile element, the I-Xe chronometry has been shown to be a promising technique for dating secondary alteration processes, that is capable of resolving age differences of a few hundred thousand years between closure times of different mineral phases from the same meteorite (e.g., *Swindle*, 1998; *Brazzle et al.*, 1999; *Pravdivtseva and Hohenberg*, 2001; *Pravdivtseva et al.*, 2001, 2003a-c; *Hohenberg et al.*, 2004). Special attention, however, must be paid to proper selection and preparation of the samples for I-Xe dating, and to its interpretation (what mineral phases or process are being dated), which should be based on detailed mineralogical study (e.g., *Krot et al.*, 1999). If samples contain more than one iodine-bearing phase and if the different mineralogical sites degas with different time-temperature profiles, stepwise pyrolysis can simulate mineral separation (*Swindle*, 1998). Whole-rock samples occasionally produce well-defined isochrons, but the results cannot be adequately interpreted if the major iodine carrier phase is unknown (e.g., *Kennedy et al.*, 1988). On the other hand, some chondritic components such as chondrules, CAIs, lithic chondritic clasts (often called "dark inclusions"), although representing mixtures of several mineral phases, can often be studied as simple objects if the major iodine carrier can be identified (e.g., *Kirschbaum*, 1988).

3. TIMESCALES AND SETTINGS FOR SECONDARY ALTERATION OF CHONDRITIC METEORITES

3.1. Timescale of Aqueous Alteration of CI Chondrites

Although CI chondrites are chemically the most primitive meteorites in that they provide the best compositional match to the solar photosphere (*Anders and Grevesse*, 1989; *Palme and*

Jones, 2003), their primary mineralogy and petrography were erased by extensive aqueous alteration at ~50-150°C on their parent body (e.g., *Richardson, 1978; McSween, 1979; Kerridge et al., 1979a,b; Bunch and Chang, 1980; Clayton and Mayeda, 1984; Zolensky et al., 1989; Endress and Bischoff, 1996; Endress et al., 1996; Leshin et al., 1997*). Subsequently, some CI chondrites experienced thermal metamorphism (e.g., *Tonui et al., 2003*). All known CI chondrites are regolith breccias consisting of various types (lithologies) of heavily-hydrated lithic fragments composed of a fine-grained phyllosilicate-rich matrix containing magnetite, sulfides, sulfates, and carbonates. The fragments are cemented by networks of secondary Ca- and Mg-sulfate veins which could be of terrestrial origin (*Gounelle and Zolensky, 2001*).

Four chemically distinct types of carbonates are found in CI chondrites: dolomite [CaMg(CO₃)₂], breunnerite [Mg(Fe,Mn)(CO₃)₂], calcite (CaCO₃) and Mg,Ca-bearing siderite (FeCO₃), with dolomite being the dominant phase (*Richardson, 1978; Fredriksson and Kerridge, 1988; Johnson and Prinz, 1993; Riciputi et al., 1994; Endress and Bischoff, 1996*). Mineralogical and isotopic (oxygen, carbon) observations suggest that the carbonates precipitated from aqueous solutions circulating on the CI parent body (e.g., *Clayton and Mayeda, 1984; Grady et al., 1988; Endress and Bischoff, 1996; Leshin et al., 2001*). Carbonates are commonly intergrown with magnetite of different textural types (platelet, framboidal, spherulitic), phosphates, and sulfides, suggesting a related paragenesis (*Endress and Bischoff, 1996*). Based on the chemical differences among dolomites within and among CI chondrites and petrographic observations of dissolution textures, composite grains etc., *Endress et al. (1996)* concluded that several episodes of aqueous alteration occurred on the CI parent body. In spite of such complexity, there have been no attempts yet to combine petrographic observations with isotopic measurements to date the different episodes of aqueous activity of the CI parent body.

3.1.1. Strontium isotope dating of CI carbonates

Strontium isotope measurements of carbonate separates from the CI chondrite Orgueil reveal that dolomite and breunnerite formed within 50 Myr after accretion of its parent body (Macdougall *et al.*, 1984; Macdougall and Lugmair, 1989). Relatively large variations of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (0.699-0.702) observed among different carbonates suggest different formation times for different types of CI carbonates (Macdougall *et al.*, 1984).

3.1.2. Chromium isotope dating of CI carbonates

Scatena-Wachel *et al.* (1984) reported $^{53}\text{Cr}^*$ corresponding to an upper limit for the $^{53}\text{Mn}/^{55}\text{Mn}$ ratio of 3.8×10^{-7} in a breunnerite grain from Orgueil, but did not draw any conclusion about the timescale of aqueous activity. Subsequently Endress *et al.* (1996) measured Cr-isotope compositions of five dolomite fragments from the CI chondrites Orgueil and Ivuna. These fragments occur between lithic clasts and are not genetically related to lithological units; they may represent debris of former carbonate veins, which were subsequently destroyed and distributed during impact-induced regolith gardening (Endress and Bischoff, 1996). All five fragments show $^{53}\text{Cr}^*$ linearly correlated with the $^{55}\text{Mn}/^{52}\text{Cr}$ ratios, indicative of *in situ* decay of ^{53}Mn (Figs. 1a,b). The data points for two fragments from Orgueil and for an Ivuna fragment plot along a line corresponding to the initial $^{53}\text{Mn}/^{55}\text{Mn}$ ratio of $(1.99 \pm 0.16) \times 10^{-6}$; the data points for two other fragments from Orgueil define a line with slope $(1.42 \pm 0.16) \times 10^{-6}$. The difference between the lines, if significant, corresponds to a time difference of 1.8 Ma. Alternatively, all five carbonates formed contemporaneously, but some of the Orgueil carbonates experienced partial isotopic equilibration of the Mn-Cr system (Endress *et al.*, 1996).

Subsequently, evidence for live ^{53}Mn in isolated carbonate grains from Orgueil (Fig. 1c; see also Fig. 1 in Hoppe *et al.*, 2004) and a CI-like clast from the Supuhee (H6) chondrite breccia

(Fig. 1d) have been reported by *Hutcheon and Phinney* (1996), *Hutcheon et al.* (1997), and *Hoppe et al.* (2001). Chromium isotope measurements in Orgueil carbonates imply $^{53}\text{Mn}/^{55}\text{Mn}$ ratio ranging from $(1.77\pm 0.15)\times 10^{-6}$ to $(3.88\pm 0.39)\times 10^{-6}$ at the time of formation of these grains. This difference in slope was interpreted as (i) it may indicate isotopic disequilibrium among carbonates subjected to several alteration events reflecting either the growth of carbonates from isotopically disparate fluids or partial Cr isotopic reequilibration, or (ii) it may have chronological significance, corresponding to an interval of ~ 4 Myr between episodes of aqueous activity. The latter is consistent with the variation in the times of formation of dolomites and breunnerites suggested from Sr isotope studies (*Macdoudal and Lugmair*, 1989). Chromium isotope measurements of carbonates in Supuhee define a correlation line with a slope corresponding to $(^{53}\text{Mn}/^{55}\text{Mn})_0$ ratio of $(8\pm 4)\times 10^{-6}$, suggesting that the aqueous activity on the CI-like parent body started earlier than recorded by the Orgueil carbonates (*Hutcheon et al.*, 1997).

The observed range in the initial $^{53}\text{Mn}/^{55}\text{Mn}$ ratios in CI carbonates [from $(1.42\pm 0.16)\times 10^{-6}$ to $(8\pm 4)\times 10^{-6}$] corresponds to an age difference of ~ 9 Ma and may represent the duration of aqueous activity of the CI parent body that started $\sim 10\pm 3$ Ma prior to differentiation of the angrite parent body.

3.1.3. Iodine-xenon isotope dating of CI magnetites

Jeffery and Anders (1970) showed that the trapped Xe resided mostly in phyllosilicates and the radiogenic ^{129}Xe in magnetite of Orgueil. Later, *Herzog et al.* (1973) and *Lewis and Anders* (1975) reported the apparent I-Xe age of the Orgueil magnetites as being ~ 7 Myr older than Shallowater and interpreted this age as the condensation time of the solar nebula. These results and interpretation are clearly inconsistent with petrographic evidence for asteroidal formation of

magnetite (e.g., *Kerridge et al.*, 1979a) and with relatively young ^{53}Mn - ^{53}Cr and $^{87}\text{Sr}/^{86}\text{Sr}$ ages of carbonates described above. Recent reexamination of the anomalous ^{129}I - ^{129}Xe age of Orgueil magnetite (fraction containing >90% magnetite) showed much later closing time of the I-Xe system, 1.9 ± 0.2 Myr older than Shallowater (Fig. 2; *Hohenberg et al.*, 2000; *Pravdivtseva et al.*, 2003b). The differences between the two studies are probably due to difficulties with the irradiation monitors in the early studies (*Hohenberg et al.*, 2000).

For a highly magnetic fraction (composed of ~14% magnetite and ~86% hydrated carbonaceous material) separated from Orgueil with a hand magnet, the I-Xe ages are 3.0 ± 0.4 Myr (*Hohenberg et al.*, 2000) and 1.5 ± 0.3 Myr (*Pravdivtseva et al.*, 2003b) younger than Shallowater, suggesting that the magnetic fraction may contain several iodine carriers recording different stages of aqueous activity on the CI parent body. This is consistent with oxygen isotopic compositions of separated components from CI chondrites (Fig. 3). In each CI chondrite, magnetite ($\Delta^{17}\text{O} = 1.3\text{‰}$ to 1.8‰) is out of oxygen isotopic equilibrium with the phyllosilicates ($\Delta^{17}\text{O} = -0.3\text{‰}$ to 0.3‰), suggesting that phyllosilicates continued to equilibrate with water as anhydrous silicates are progressively altered to phyllosilicates, whereas isotope exchange between magnetite and fluid was kinetically slow (*Rowe et al.*, 1994). Carbonates ($\Delta^{17}\text{O} = 0.3\text{‰}$ to 0.5‰) are in isotopic equilibrium with phyllosilicates (*Leshin et al.*, 2001), suggesting precipitation from a fluid of similar oxygen isotopic composition.

3.2. Timescale of Aqueous Alteration of the Polymict Chondrite Breccia Kaidun: Evidence from Chromium Isotope Compositions of Carbonates

Kaidun is a polymict chondrite breccia containing lithic clasts of the C1, CM-like, CR-like, CV, R, EH, and EL chondrites (*Clayton and Mayeda*, 1999; *Zolensky and Ivanov*, 2003). Most

clasts have been extensively altered at ~250-450°C by hydrothermal fluids that resulted in formation of phyllosilicates, and carbonate- and phyllosilicate-filled veins (*Johnson and Prinz, 1993; Weisberg et al., 1994; Zolensky et al., 1996*). All lithic clasts contain carbonates, with calcite being the dominant phase; dolomite is less abundant (*Johnson and Prinz, 1993; Weisberg et al., 1994*). Calcite occurs within altered chondrules, CAIs and mineral fragments, and as fragments dispersed throughout the matrix and in veins. The veins occur along the boundaries between lithic clasts, suggesting some calcite formed after agglomeration of the Kaidun breccia.

Chromium isotopic compositions of five calcite and one dolomite grain from three different lithologies (CR-like, CM1, and C1) measured by *Hutcheon et al. (1999)* are plotted in Figure 4a. The slope of the correlation line on a ^{53}Mn - ^{53}Cr evolution diagram corresponds to an initial $^{53}\text{Mn}/^{55}\text{Mn}$ ratio of $(9.4\pm 1.6)\times 10^{-6}$, suggesting nearly contemporaneous formation of carbonates in the lithologies studied ($\Delta t_{\text{LEW}} \sim 10.8\pm 1$ Myr).

3.3. Timescale of Aqueous Alteration of CM chondrites

The CM carbonaceous chondrites are a diverse group of petrologic type 1-2 meteorites that experienced low-temperature (~0-25°C) aqueous alteration to various degrees in an asteroidal setting that resulted in formation of a variety of secondary phases, including phyllosilicates, magnetite, Fe,Ni-sulfides, and carbonates (e.g., *Kerridge and Bunch, 1979; Zolensky and McSween, 1988; Zolensky et al., 1993; Brearley and Jones, 1998*). Some CM chondrites [e.g., Begica-7904, Yamato (Y) 86720] subsequently experienced thermal metamorphism and partial dehydration (e.g., *Tomeoka et al., 1989; Tomeoka, 1990; Ikeda, 1992; Clayton and Mayeda, 1999*).

Detailed mineralogical and isotopic studies of CM carbonates revealed their complex formation history, involving periods of dissolution and reprecipitation due to interactions with fluids of different compositions (e.g., *Zolensky et al.*, 1989; *Johnson and Prinz*, 1993; *Riciputi et al.*, 1994; *Brearley et al.*, 1999, 2001; *Brearley and Hutcheon*, 2000, 2002; *Benedix et al.*, 2003). Carbonates occur in fine-grained rims around chondrules and mineral fragments and within altered CAIs (e.g., *Bunch and Chang*, 1980). There is no common association of carbonates with other phases in CM chondrites, although textural observations suggest that carbonates must have coprecipitated with phyllosilicates, magnetite, and tochilinite (e.g., *Kerridge and Bunch*, 1979; *Bunch and Chang*, 1980; *Barber*, 1981; *Mackinnon et al.*, 1984; *Johnson and Prinz*, 1993). However, oxygen isotope compositions of carbonates (*Clayton and Mayeda*, 1984; *Brearley et al.*, 1999; *Benedix et al.*, 2003), magnetite (*Rowe et al.*, 1994), and phyllosilicate-rich matrix (*Clayton and Mayeda*, 1984) in the CM chondrite Murchison (Fig. 3) indicate that the phyllosilicates are not in isotope equilibrium with carbonates and magnetite. It is suggested that carbonates and magnetites precipitated from an isotopically evolving water reservoir, prior to formation of phyllosilicates (*Rowe et al.*, 1994; *Brearley et al.*, 1999; *Benedix et al.*, 2003). Timing of aqueous activity on the CM parent body remains poorly constrained and is largely based on a limited number of chromium isotope measurements in carbonates (*Brearley and Hutcheon*, 2000; *Brearley et al.*, 2002).

3.3.1. Chromium isotope dating of CM carbonate formation

In the relatively weakly-altered and virtually unbrecciated CM chondrite Y791198 calcite is the only carbonate present. Calcite grains show complex zoning indicating periods of dissolution and reprecipitation (*Brearley et al.*, 2001). Three out of six calcite grains analyzed for Cr isotopic compositions showed the presence of $^{53}\text{Cr}^*$ correlated with the respective $^{55}\text{Mn}/^{52}\text{Cr}$ ratios,

indicating *in situ* decay of ^{53}Mn . The slope of the correlation line on a ^{53}Mn - ^{53}Cr evolution diagram corresponds to an initial $^{53}\text{Mn}/^{55}\text{Mn}$ ratio of $(8.7\pm 1.5)\times 10^{-6}$ (Fig. 4b).

In the heavily-altered CM1 chondrite Allan Hills (ALH) 84034 both calcite and dolomite are present; calcite is much less abundant (*Brearley and Hutcheon, 2000*). The two phases always occur separately, except within altered CAIs where they can coexist. Dolomites are commonly intergrown with serpentines and pentlandite; calcites are inclusion-free. One of the dolomite grains exhibits resolvable $^{53}\text{Cr}^*$ corresponding to an initial $^{53}\text{Mn}/^{55}\text{Mn}$ ratio of $(5.0\pm 1.5)\times 10^{-6}$ (Fig. 4c).

The observed range in the initial $^{53}\text{Mn}/^{55}\text{Mn}$ ratios in CM carbonates [from $(1.3\pm 0.6)\times 10^{-5}$ to $(5.0\pm 1.5)\times 10^{-6}$] corresponds to an age difference of ~ 5 Ma and may represent duration of aqueous activity of the CM parent body that started $\sim 12.5\pm 2.5$ Ma prior to differentiation of the angrite parent body (*Lugmair and Shukolyukov, 1998*).

3.4. Timescale of *in situ* Aqueous Alteration of the Ungrouped Carbonaceous Chondrite MAC88107: Evidence from Petrographic Observations and Chromium Isotope Compositions of Secondary Fayalite

The ungrouped carbonaceous chondrite MacAlpine Hills (MAC) 88107 has a bulk chemical composition intermediate between CO and CM chondrites, and O-isotopic composition similar to CO-CV-CK chondrites (*Clayton and Mayeda, 1999*). In contrast to CK and most CO chondrites, MAC88107 shows no evidence for thermal metamorphism; its thermoluminescence properties (*Sears et al., 1991*) suggest low petrographic type (3.0-3.1). Chondrules and CAIs are surrounded by continuous fine-grained, accretionary rims, indicating that the meteorite largely escaped post-accretionary brecciation (*Krot et al., 2000a*).

The meteorite experienced a small degree of *in situ* alteration that resulted in formation of saponite, serpentine, magnetite, pentlandite, fayalite, and hedenbergite (Krot *et al.*, 2000a). We emphasize that similar secondary phases are observed in the Bali-like oxidized CV chondrites, where their origin remains controversial (see below). Because, MAC88107 may provide a clue for understanding the origin of secondary mineralization in CV chondrites, it is discussed in detail in this chapter. Fayalite (Fa₉₀₋₁₀₀) and hedenbergite (~Fs₅₀Wo₅₀) occur as veins, which start at the opaque nodules in the chondrule peripheries, crosscut fine-grained rims and either terminate at the boundaries with the neighboring fine-grained rims or continue as layers between these rims (Fig. 5a,b). Fayalite also overgrows isolated forsteritic (Fa₁₋₅) and fayalitic (Fa₂₀₋₄₀) olivine grains without any evidence for Fe-Mg interdiffusion (Fig. 5c), and replaces magnetite-sulfide grains (Fig. 5d). All textural varieties of fayalite are compositionally similar and characterized by high MnO content (0.4-0.85 wt%) and nearly complete absence of Cr₂O₃.

Based on the petrographic observations and thermodynamic analysis of phase relations in the Si-Fe-Ca-O-H system, Krot *et al.* (2000a) concluded that phyllosilicates, magnetite, pentlandite, fayalite, and hedenbergite in MAC88107 formed during low-temperature (~150-200°C) alteration in the presence of aqueous solution capable of transporting Si, Fe, Ca, Mn, and Mg. Because most fayalite grains in MAC88107 are too small (<10 µm) for Mn-Cr-isotope study by an ion microprobe, the Cr isotope compositions were measured only for a coarse-grained fayalite replacing a magnetite-sulfide nodule (Fig. 5d) and adjacent matrix. Both analyses of the fayalite grain show large ⁵³Cr* correlated with the respective ⁵⁵Mn/⁵²Cr ratios, indicative for the *in situ* decay ⁵³Mn (Fig. 6a). The slope of the correlation line fitted to the data, passing through the normal Cr isotope composition of matrix (δ⁵³Cr = 0) at Mn/Cr ≅ 1, corresponds to the initial ⁵³Mn/⁵⁵Mn ratio of (1.58±0.26)×10⁻⁶ at the time the fayalites formed (Δt_{LEW} = -1.25±0.83 Ma).

3.5. CV Chondrites and Settings of Their Alteration

The CV carbonaceous chondrites are currently subdivided into the reduced (CV_{red}) and two oxidized subgroups, Allende-like (CV_{oxA}) and Bali-like (CV_{oxB}) (McSween, 1977; Weisberg and Prinz, 1998), which largely reflect their complex alteration history and may represent different lithological varieties of the same asteroidal body (Krot *et al.*, 1998a).

The CV_{oxB} chondrites (e.g., Kaba, Bali) experienced aqueous alteration resulting in replacement of primary minerals in chondrules, CAIs, and AOAs by secondary phyllosilicates, magnetite, Fe,Ni-sulfides, Fe,Ni-carbides, fayalite, salite-hedenbergite pyroxenes ($Fs_{10-50}Wo_{45-50}$), and andradite. Their matrices largely consist of the secondary minerals, including concentrically-zoned nodules of Ca,Fe-pyroxene and andradite, coarse ($>10\ \mu m$) grains of nearly pure fayalite ($>Fa_{90}$), abundant phyllosilicates, and very fine-grained ($<1-2\ \mu m$) ferrous olivine ($\sim Fa_{50}$) (Fig. 7a).

The CV_{oxA} chondrites (e.g., Allende, ALH84128) are more extensively altered than the CV_{oxB} , but contain very minor phyllosilicates (this alteration is often referred as iron-alkali metasomatism). Chondrules and refractory inclusions in the CV_{oxA} chondrites contain secondary nepheline, sodalite, Ca,Fe-pyroxenes, andradite, Fe,Ni-sulfides, magnetite, Ni-rich metal, and ferrous olivine. Their matrices are coarser-grained than those in the CV_{oxB} and largely consist of Ca,Fe-pyroxene±andradite nodules, lath-shaped ferrous olivine ($\sim Fa_{50}$), and nepheline (Fig. 7c).

The CV chondrite Mokoia is a complex breccia containing clasts of the CV_{oxA} and CV_{oxB} lithologies and heavily-metamorphosed oxidized chondritic clasts (Krot *et al.*, 1998a). The CV_{oxA} clasts experienced aqueous alteration that overprints "anhydrous" Allende-like alteration (Kimura and Ikeda, 1998). Some oxidized CVs (e.g., MET00430) are mineralogically intermediate between the CV_{oxB} and CV_{oxA} chondrites (Fig. 7b; Krot *et al.*, 2004a).

The CV_{red} chondrites Efremovka and Leoville experienced alteration similar to that of CV_{oxA}, but to a smaller degree. The reduced CV chondrite breccia Vigarano contains clasts of the CV_{oxB} and CV_{oxA} materials (Krot *et al.*, 2000b); the reduced portion experienced aqueous alteration resulting in formation of phyllosilicates and magnetite.

In addition to the oxidized and reduced CV subgroups, CV chondrites contain dark inclusions which are chemically and petrographically similar to their host meteorites (Fig. 7d), but appear to have experienced more extensive alteration (e.g., Fruland *et al.*, 1978; Kurat *et al.*, 1989; Johnson *et al.*, 1990; Buchanan *et al.*, 1997; Krot *et al.*, 1997a, 1998a, 1999, 2001).

3.5.1. Nebular vs. Asteroidal alteration of CV chondrites

The origin of secondary mineralization in CV chondrites remains controversial; nebular and asteroidal settings have been proposed. According to the nebular models (Palme and Wark, 1988; Weisberg and Prinz, 1998), chondrules and refractory inclusions in the CV_{oxA} were exposed to a highly oxidized nebular gas resulting in their alteration; matrix minerals directly condensed from this gas. This model is, however, inconsistent with (a) the presence of poorly-graphitized carbon (PGC) and pentlandite inclusions in matrix olivine (Brearley, 1999), (b) the lack of volatility-controlled rare earth element (REE) patterns in matrix Ca,Fe-pyroxenes and andradite (Brearley and Shearer, 2000), (c) the large mass-dependent fractionation of oxygen isotopes ($\delta^{18}\text{O} \sim 20\text{‰}$) in secondary fayalite, magnetite, Ca,Fe-rich pyroxenes, and andradite (Krot *et al.*, 2000c; Choi *et al.*, 2000; Cosarinsky *et al.*, 2003), and (d) the thermodynamic analysis of condensation of ferrous olivine (Grossman and Fedkin, 2003).

According to the asteroidal models (Krot *et al.*, 1995, 1997, 1998a,b, 2004a; Kojima and Tomeoka, 1996), CV chondrites experienced fluid-assisted thermal metamorphism to various degrees, which resulted in mobilization of Ca, Si, Fe, Mg, Mn, Na, and S, and replacement of

primary phases in chondrules, CAIs and matrices by secondary minerals. It was originally suggested that secondary ferrous olivine in CV chondrites formed by dehydration of phyllosilicates during thermal metamorphism (*Kojima and Tomeoka, 1996; Krot et al., 1995, 1997a*). This mechanism, however, appears to be inconsistent with the lack of mass-dependent fractionation of oxygen isotopes in bulk CV chondrites (*Clayton and Mayeda, 1999*), which is expected for extensively aqueously-altered and dehydrated meteorites (e.g., metamorphosed CI/CM). Recently, *Krot et al. (2004a)* concluded that ferrous olivine in CV chondrites formed by several mechanisms during fluid-assisted metamorphism, including replacement of opaque nodules, magnesian olivine and pyroxene, direct precipitation from a supersaturated fluid, and, possibly by dehydration of phyllosilicates.

3.5.2. Settings and timescale of secondary alteration of the CV_{oxB} chondrites

There are several lines of evidence suggesting that the secondary minerals in the CV_{oxB} formed during relatively low-temperature aqueous alteration of the CV asteroidal body, rather than by high-temperature gas-solid reactions in the solar nebula. (a) The secondary minerals occur in all CV_{oxB} chondritic components, including chondrules, CAIs, AOAs, and matrices. (b) Fine-grained rims around chondrules are commonly crosscut by fayalite-bearing veins that start at the opaque nodules in the chondrule peripheries (Figs. 8a,b). (c) Fayalite replacing magnetite-sulfide nodules in type I chondrules (Figs. 8c,d) show large mass-dependent fractionation of oxygen isotopes and contain sulfide inclusions (Fig. 9a), suggesting low-temperature formation. (d) Euhedral fayalite grains of variable compositions overgrow forsterite grains of AOAs without any evidence for Fe-Mg interdiffusion in the neighboring forsterite grains, suggesting precipitation from a low-temperature fluid of variable chemical composition (Fig. 8e). Occasionally, ferrous olivine pseudomorphs chondrule phenocrysts (Fig. 8d) supporting the

presence of aqueous solutions during the alteration. (e) Low Al contents in secondary Ca,Fe-pyroxenes, indicating large Ca/Al fractionation during their formation, is inconsistent with their high-temperature condensation origin (both Ca and Al are refractory lithophile elements of similar volatility and are not expected to be fractionated from each other during condensation). These observations and thermodynamic analysis of phase relations in the Si-Fe-Ca-O-H system (Krot *et al.*, 1998a) suggest that secondary minerals in CV_{oxB} chondrites in the presence of aqueous solutions capable of transporting Si, Fe, Ca, Mn, and Mg.

Petrographic observations of type I chondrules (Hua and Buseck, 1995; Krot and Todd, 1998; Krot *et al.*, 1998a,b) suggest the following sequence of secondary mineral formation. Magnetite and Fe,Ni-sulfides replacing Fe,Ni-nodules formed first. Phyllosilicates replacing chondrule mesostases and phenocrysts formed either subsequently or contemporaneously with magnetite. Fayalite, Ca,Fe-pyroxenes and andradite replace magnetite and coexist with phyllosilicates, possibly indicating contemporaneous formation of these phases; occasionally, fayalite is corroded by Ca,Fe-pyroxenes.

3.5.2.1. Manganese-chromium isotope dating of secondary fayalite in Kaba and Mokoia

High MnO contents (up to 1.5 wt%) in secondary fayalite and nearly complete absence of chromium (Mn/Cr ratios range up to 2×10^6) favor chromium isotope measurements of fayalite to constrain its crystallization time (Hutcheon *et al.*, 1998; Hua *et al.*, 2003, 2004). Hutcheon *et al.* (1998) measured Cr isotope compositions of six fayalite grains replacing magnetite-sulfide nodules within type I chondrules from Mokoia. Hua *et al.* (2002, 2004) analyzed Cr isotope compositions of twelve fayalite grains associated with magnetite and sulfides in Kaba matrix. Fayalite grains in both textural occurrences have large $^{53}\text{Cr}^*$ correlated with $^{55}\text{Mn}/^{52}\text{Cr}$ ratios, indicative for *in situ* decay of ^{53}Mn , which define similar initial $^{53}\text{Mn}/^{55}\text{Mn}$ ratios of

$(2.32 \pm 0.18) \times 10^{-6}$ and $(2.28 \pm 0.37) \times 10^{-6}$, respectively ($\Delta_{\text{ILEW}} = \sim 3.0 \pm 0.7$ Myr) respectively (Figs. 6b,c).

3.5.2.2. Iodine-xenon dating of magnetite and phyllosilicates formation in Kaba and Bali

Pravdivtseva and Hohenberg (2001) measured Xe isotope compositions of magnetic fractions separated with a hand magnet from fine-crushed Kaba, Bali, and Mokoia. The ^{128}Xe and ^{129}Xe release profiles in Kaba and Bali suggest one major iodine carrier in magnetic separates that yield well-defined isochrons in temperature ranges of 1400-1750°C and 1450-1950°C, respectively (Figs. 10a,d). The isochrons correspond to closure time of the I-Xe system in the Kaba and Bali magnetite of 4.2 ± 0.3 Ma and 7.9 ± 0.2 Ma relative to the Shallowater internal reference standard, respectively.

The ^{128}Xe and ^{129}Xe release profiles at 1400-1900°C of the Mokoia magnetic fraction suggest multiple iodine carrier; no isochron was obtained. These results are consistent with the complex brecciated nature and multistage alteration history of this meteorite (*Krot et al.*, 1998a; *Kimura and Ikeda*, 1998).

Two non-magnetic fractions hand picked from coarsely crushed samples of Kaba and Bali and tentatively identified (using EDS) as enstatite and a mixture of plagioclase-rich mesostasis and Al-rich phyllosilicates were measured for Xe isotope compositions (*Pravdivtseva et al.*, 2001). Considering very fine scale of primary and secondary mineral intergrowths in the CV_{oxB} chondrules, it is difficult to expect good mineral separation, and the results should be treated cautiously. The enstatite separates define precise high-temperature isochrons from $\sim 1400^\circ\text{C}$ to $\sim 1800^\circ\text{C}$ with similar I-Xe ages: -2.0 ± 0.8 Ma for Kaba and -2.1 ± 0.7 Ma for Bali, relative to Shallowater internal standard (Figs. 10b,e). The mixture of plagioclase-rich mesostasis and Al-rich phyllosilicates yield lower temperature isochrons corresponding to I-Xe ages of 8.9 ± 0.7 Ma

and 9.0 ± 0.8 Ma, for Kaba and Bali, respectively (Figs. 10c,f). These ages are systematically younger than the corresponding magnetite ages and may suggest that either magnetite formation predates formation of phyllosilicates or that I-Xe isotope closure in magnetite occurred prior to that in phyllosilicates. The overall I-Xe data suggest that the aqueous alteration on the CV_{oxB} parent body lasted for at least 10 Ma.

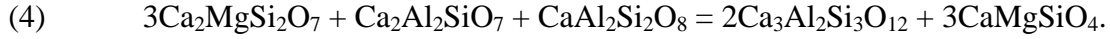
3.5.3. Settings and timescale of alteration of the CV_{oxA} chondrites

There are several lines of evidence suggesting that iron-alkali metasomatic alteration of the CV_{oxA} chondrites resulted from fluid-assisted thermal metamorphism of the CV asteroidal body (e.g., *Krot et al.*, 1998a), rather than from high-temperature gas-solid reactions in the solar nebula (e.g., *Palme and Wark*, 1988). (a) The secondary minerals occur in all CV_{oxA} chondritic components, including chondrules, CAIs, AOAs, and matrices (e.g., *Hashimoto and Grossman*, 1987; *MacPherson et al.*, 1988; *Krot et al.*, 1995) and show evidence for *in situ* formation (e.g., veins, rims, chondrule pseudomorphs; Fig. 11) (*Krot et al.*, 1997a, 1998a,b, 2001; *MacPherson and Krot*, 2002). (b) Oxygen isotope compositions of the secondary Ca,Fe-pyroxenes, andradite, and wollastonite in matrix and rims around CAIs plot parallel to terrestrial fractionation line at $\Delta^{17}\text{O} \sim -2.5\text{‰}$ with a large range in $\delta^{18}\text{O}$ ($\sim 20\text{‰}$) (Fig. 9b), comparable to the range reported for the secondary magnetites and fayalites in the CV_{oxB} chondrites (Fig. 9a), suggesting low-temperature formation. This mechanism is also consistent with the presence of sulfide inclusions (Fig. 11b) and lack of volatility-controlled REE patterns in Ca,Fe-pyroxenes and andradite in the Allende matrix (*Brearley and Shearer*, 2000). (c) Secondary ferrous olivine replacing low-Ca pyroxene phenocrysts in type I chondrules coexists with talc and amphibole (*Brearley*, 1999), suggesting that Fe was transported by low-temperature aqueous solutions (*Krot et al.*, 2004a), rather than by high-temperature gas phase (*Dohmen et al.*, 1998).

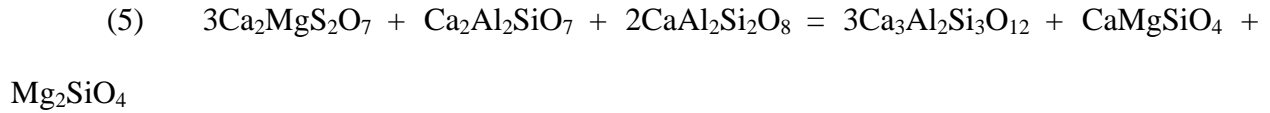
Although there are many textural and mineralogical similarities in secondary alteration of the CV_{oxB} and CV_{oxA} chondrites (Figs. 8, 11a-d), the latter are more extensively altered and contain secondary ferrous olivine (Fa₄₀₋₆₀), nepheline and sodalite instead of fayalite (Fa₉₀₋₁₀₀) and phyllosilicates. There are also some difference in $\Delta^{17}\text{O}$ values of the secondary phases (fayalite, magnetite, and Ca,Fe-rich silicates) in the CV_{oxB} (~-0.6‰) and CV_{oxA} chondrites (-2.6‰) (Figs. 9a,b). Secondary fayalites in the intermediate CV_{oxA-B} meteorites (e.g., MET00430) show inverse compositional zoning (Figs. 7b, 11e) and evidence for dissolution of fayalite and precipitation of more forsteritic olivine (Fig. 11f). These observations may indicate that the CV_{oxA} experienced alteration at higher temperatures than the CV_{oxB}.

Petrographic observations on type I chondrules in the CV_{oxA} (Figs. 11a-d; *Krot et al.*, 1998a,b) suggest the following sequence of secondary mineral formation. Magnetite and Fe,Ni-sulfides replacing Fe,Ni-nodules formed first. Ferrous olivine and Ca,Fe-pyroxenes formed later; they preferentially replace magnetite of the nodules and contain abundant inclusions of Fe,Ni-sulfides (Figs. 11a-c). Ferrous olivine also replaces low-Ca pyroxene phenocrysts and overgrows or replaces forsteritic olivine phenocrysts (Figs. 10c,d; *Krot et al.*, 1997a). Nepheline and sodalite replace chondrule mesostasis and may have formed prior to, contemporaneously or after ferrous olivine (e.g., *Kimura and Ikeda*, 1995).

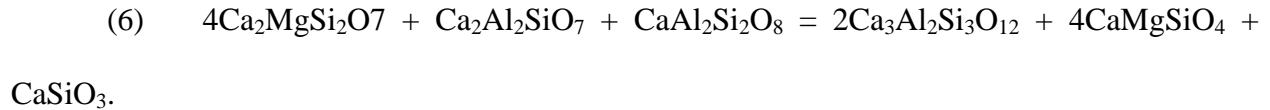
In addition to iron- and alkali-rich minerals in the CV_{oxA} chondrites, coarse-grained CAIs in Allende contain secondary grossular, monticellite, wollastonite, and forsterite which typically replace melilite-anorthite assemblages (*Hutcheon et al.*, 1978; *Hutcheon and Newton*, 1981; *Wark*, 1987; *Krot et al.*, 2004b). Based on petrographic observations and thermodynamic analysis, *Hutcheon and Newton* (1981) concluded that grossular and monticellite formed during a prolonged heating in the solar nebula at ~950 K via the closed-system reaction:



Krot et al. (2004b) concluded instead that other closed-system reactions took place:



and



Although under equilibrium conditions these reaction occur below 950 K, the common presence of unaltered melilite-anorthite intergrowths in the Allende Type C CAIs implies the lack of equilibrium (i.e. temperature estimates should be considered with caution).

3.5.3.1. Aluminum-magnesium isotope dating of secondary alteration of the CV CAIs

Secondary minerals (nepheline, sodalite, grossular) in the CV CAIs generally show no evidence for $^{26}\text{Mg}^*$ suggesting that the alteration took place at least several half-lives of ^{26}Al after the formation of the primary phases typically having canonical $^{26}\text{Al}/^{27}\text{Al}$ ratios of $\sim 5 \times 10^{-5}$ (e.g., *Hutcheon and Newton*, 1981). The only Allende CAI with excesses of ^{26}Mg in secondary nepheline and sodalite is a fine-grained spinel-rich inclusion analyzed by *Brigham et al.* (1986). The observed $^{26}\text{Mg}^*$ corresponds to an initial $^{26}\text{Al}/^{27}\text{Al}$ ratio of $(6-7) \times 10^{-5}$. *MacPherson et al.* (1995) interpreted these data as evidence for an early, nebular formation of the secondary minerals that continued over an extended (several Ma) period of time. However, taking into account the low $^{27}\text{Al}/^{24}\text{Mg}$ ratios in the analyzed minerals, the anomalously high initial $^{26}\text{Al}/^{27}\text{Al}$ ratio inferred for this CAI, and the clear evidence for metamorphic redistribution of Mg isotopes in the Allende CAIs (e.g., *Yurimoto et al.*, 2000), it seems more likely that this "isochron"

resulted from Mg-isotopic exchange between the primary and secondary minerals of the CAI and does not have a chronological meaning (see also Fig. 2 in *MacPherson et al.*, 1995).

3.5.3.2. Iodine-xenon dating of secondary alteration of CAIs and chondrules in Allende

I-Xe isotope data for the coarse-grained and fine-grained CAIs in Allende, which experienced iron-alkali metasomatic alteration, encompass a spread of ≥ 10 Ma, supporting an asteroidal setting of alteration (*Swindle et al.*, 1988). The strong correlation of iodine with chlorine in two fine-grained CAIs analyzed by *Kirschbaum* (1988) together with the fact that sodalite is the only significant chlorine-bearing mineral in these CAIs, verified sodalite as the major iodine-carrier phase.

Recent xenon isotope measurements (*Pravdivtseva et al.*, 2003b) showed that heavily-altered fine-grained CAIs in Allende define isochrons with ages between 3.1 ± 0.2 and 3.7 ± 0.2 Ma younger than Shallowater (Figs. 12a-c). The CAIs have nearly identical release profiles for radiogenic ^{129}Xe and ^{128}Xe , suggesting the same iodine carrier for both (probably sodalite; *Kirschbaum*, 1988).

Although Allende chondrules often contain large fractions of radiogenic Xe, and an I-Xe association suggestive chronometry, they rarely yield isochrons that are well-defined at the level of precision provided by the isotopic data. Among nine chondrules studied by *Swindle et al.* (1983), eight have a pattern of increasing model age with each incrementally increased temperature step. This was attributed to relatively slow cooling ($\sim 10\text{-}20^\circ\text{C}/\text{Ma}$ using the lower release temperatures ($600\text{-}1100^\circ\text{C}$) or of $50\text{-}300^\circ\text{C}/\text{Ma}$ using release temperatures above 1300°C) or the monotonic (with release temperature) relaxation of other conditions during thermal metamorphism or alteration (*Swindle et al.*, 1983; *Nichols et al.*, 1990). One chondrule gave a well-defined isochron with an apparent age of 0.53 ± 0.15 Ma younger than the Bjurböle

whole rock age standard (Swindle *et al.*, 1983), and the I-Xe ages of four different chondrules gave ages ranging from -0.37 ± 0.16 Ma to 1.54 ± 0.07 Ma, relative to Bjurböle (Nichols *et al.*, 1990). Four coarse-grained chondrule rims tended to be slightly older than the interiors, but these rims were separated from a different set of chondrules, and the only chondrule/rim pair combination yielded concordant ages.

3.5.4. Setting and I-Xe dating of alteration of the CV dark inclusions

Dark inclusions in Allende experienced similar secondary alteration to their host meteorite, but to a higher degree (Figs. 13a,b). The very heavily-altered dark inclusions consist almost entirely of secondary ferrous olivine, Ca,Fe-pyroxenes, andradite, nepheline, sodalite, and Fe,Ni-sulfides, and, if not brecciated, are surrounded by continuous, multilayered Ca,Fe-rich rims (Fig. 14) composed of Ca,Fe-pyroxenes, andradite, \pm wollastonite, \pm kirschsteinite. The outermost layer of the rims is often intergrown with chondrule fragments and matrix olivine of the Allende host (Fig. 13f in Krot *et al.*, 1998a; Fig. 10 in Krot *et al.*, 2001). Some of the dark inclusions are crosscut by multiple Ca,Fe-rich veins (Fig. 14a), which are mineralogically similar to the Ca,Fe-rich rims and often connected to them. The outer portions of the rimmed dark inclusions are depleted in Ca, whereas the Allende matrix just outside the rims contain abundant Ca,Fe-rich silicate inclusions (Figs. 14a,b). Oxygen isotope compositions of Ca,Fe-rich silicates within and around dark inclusions in Allende (Fig. 9c) plot parallel to the terrestrial fractionation line at $\Delta^{17}\text{O} \sim -2\text{‰}$ with a large range in $\delta^{18}\text{O}$ ($\sim 30\text{‰}$), suggesting low temperature formation of these minerals (Krot *et al.*, 2000c).

Based on these observations and thermodynamic analysis of phase relations in the Si-Fe-Ca-O-H system, Krot *et al.* (2001) concluded that the rimmed dark inclusions in Allende experienced at least two-stages of alteration in the presence of aqueous solutions. During an

early stage of the alteration, which took place in an asteroidal setting, but not in the current location of the dark inclusions, chondrule silicates were replaced by secondary ferrous olivine, nepheline, and sodalite. Calcium lost from the chondrules was redeposited as Ca,Fe-pyroxene-andradite veins and nodules in the dark inclusion matrices. The second stage of alteration took place *in situ*, during the alteration of the Allende host, and resulted in mobilization of Ca from the dark inclusions and its redeposition as Ca,Fe-rich rims around the dark inclusions and as Ca,Fe-rich nodules in the neighboring matrix of Allende.

Xenon isotope compositions were measured in bulk samples of seventeen Allende dark inclusions (*Pravdivtseva et al.*, 2003b). All dark inclusions yielded similar release profiles with two major peaks, suggesting two major iodine carriers (sodalite, and possibly Ca,Fe-pyroxenes or nepheline), and well-defined I-Xe isochrons (Figs. 12d-f) with ages ranging from 0.5 ± 0.3 to 2.8 ± 0.3 Ma older than the Shallowater internal standard (Table 1). In contrast, three heavily-altered fine-grained CAIs in Allende yielded well-defined isochrons with ages 3.1 ± 0.2 , 3.0 ± 0.2 , and 3.7 ± 0.2 Ma younger than Shallowater (*Pravdivtseva et al.*, 2003b). The I-Xe ages of the dark inclusions were interpreted as the time of their early alteration prior incorporation into Allende. The younger I-Xe ages of the fine-grained spinel-rich CAIs may reflect hydrothermal alteration of the Allende host, which could have occurred contemporaneously with the second stage of alteration of the Allende dark inclusions. The lack of evidence for the disturbance of I-Xe system in the Allende dark inclusions suggests that fluid responsible for the alteration of the Allende CAIs must have been in equilibrium with the I- and Xe-bearing phases of the dark inclusions, so the latter were not affected by the second stage of alteration.

Dark inclusions in the CV_{red} chondrites Efremovka, Leoville, and Vigarano experienced different styles of aqueous alteration to various degrees (Figs. 13c-f) that resulted in formation of

ferrous olivine, andradite, magnetite, and phyllosilicates (*Kracher et al.*, 1985; *Tomeoka and Kojima*, 1998; *Brearley*, 1998; *Krot et al.*, 1999). The presence of aqueous solutions during the alteration is supported by the textural observations (e.g., chondrule pseudomorphs), the presence of minor phyllosilicates (*Krot et al.*, 1999), and bulk oxygen isotope compositions, which on a three oxygen isotope plot deviate to the right from the CCAM line (*Clayton and Mayeda*, 1999). Xenon isotope compositions were measured in bulk samples of six dark inclusions from the reduced CVs (*Swindle et al.*, 1998; *Krot et al.*, 1999; *Pravdivtseva et al.*, 2003c). The iodine carriers in the dark inclusions have not been identified; phyllosilicates and magnetite are two possible candidates. The I-Xe ages of the dark inclusions range from -4.9 ± 1.8 to 9.5 ± 2.3 Ma relative to Shallowater and are generally younger than those of the Allende dark inclusions (Table 1). For the Efremovka dark inclusions, there is a correlation between the degree of alteration and I-Xe closure times: E39 and E80 are more altered than E53 and show an apparent closure time ~ 4 to 6 ± 2 Ma later than E53 (Fig. 15).

In spite of the different degrees and styles of alteration of dark inclusions in the reduced and oxidized CV chondrites, all of them require "sub-planetary" trapped Xe, which has been interpreted as a result of shock or thermal metamorphism that occurred after precipitation of iodine host, while Xe and some I were still in solution (*Hohenberg et al.*, 2003). This interpretation is generally consistent with the aqueous alteration - dehydration model proposed for the dark inclusions (e.g., *Kojima and Tomeoka*, 1996) and with shock metamorphism features observed in some of the dark inclusions (e.g., lineation of chondrule pseudomorphs; Fig. 1 in *Krot et al.*, 1999).

To summarize, the I-Xe ages of the CV dark inclusions, which probably represent fragments of the CV asteroidal body, span ~14 Ma, suggesting a long period of aqueous alteration on the CV parent body.

3.6. Timescale of Aqueous Alteration of Ordinary Chondrites

The effects of aqueous alteration are best documented in chondrules and matrices of the type 3 ordinary chondrites Semarkona (LL3.0), Bishunpur (LL3.1), Krymka (LL3.1), Parnallee (LL3.4), Chainpur (LL3.4), and Tieschitz (H/L3.6) (*Hutchison et al.*, 1987, 1998; *Alexander et al.*, 1989a,b; *Bridges et al.*, 1997; *Krot et al.*, 1997b; *Keller*, 1998; *Choi et al.*, 1998; *Grossman et al.*, 2000, 2002). This alteration must have occurred in an asteroidal setting and resulted in formation of secondary phyllosilicates, magnetite, maghemite, Fe,Ni-carbides, calcite, Ni-bearing sulfides, ferrous olivine, and alkali-rich secondary phases. Chondrules in some of the altered ordinary chondrites were dated using I-Xe systematics (*Swindle et al.*, 1991a,b; *Ash et al.*, 1995).

In Semarkona, evidence for aqueous alteration in an asteroidal setting include (a) the large range in mass-dependent fractionation of oxygen isotope compositions of magnetite grains ($\delta^{18}\text{O} \sim 13\%$), indicative of Rayleigh fractionation as a result of growth in the presence of a limited water reservoir (*Choi et al.*, 1998), (b) the presence of carbide-magnetite-sulfide veins crosscutting fine-grained rims around chondrules (*Krot et al.*, 1997b), (c) the presence of phyllosilicates in the chondrules and matrix (*Hutchison et al.*, 1987; *Alexander et al.*, 1989a,b), (d) the presence of bleached chondrules and evidence for removal of chondrule mesostasis by dissolution, (e) the elevated D/H ratios in the bleached chondrules and matrix of Semarkona, suggesting exchange with an isotopically similar reservoir, most likely aqueous solution (*Grossman et al.*, 2000). *Swindle et al.* (1991a) observed a range of > 10 Ma in apparent I-Xe

isotope ages (from -4.4 ± 2.9 Ma to 5.4 ± 0.5 Ma, relative to Shallowater) for seventeen chondrules analyzed in Semarkona (Table 2; Fig. 16). The oldest I-Xe ages were attributed to chondrule formation, whereas the younger ages to aqueous alteration. Taking into account the petrographic evidence for multistage aqueous alteration of the Semarkona chondrules (e.g., *Grossman et al.*, 2000) and the possible presence of several iodine carriers (e.g., magnetite, phyllosilicates), we suggest instead that the entire range of I-Xe ages may reflect duration of aqueous alteration on the LL asteroidal body.

The H/L3.6 chondrite Tieschitz contains secondary nepheline, albite, and unidentified hydrous (?) phases that precipitated from a halogen-bearing aqueous fluid in interchondrule voids and replaced chondrule mesostasis leached out by the fluid (*Hutchison et al.*, 1998). Based on the evidence for partial resetting of the Sm-Nd and K-Ar systems at ~ 2 Ga (*Turner et al.*, 1978; *Krestina et al.*, 1996), *Hutchison et al.* (1998) speculated that aqueous activity on the Tieschitz parent body occurred ~ 2 Ga ago. However, the I-Xe ages of the Tieschitz chondrules (*Nichols et al.*, 1991) do not support this hypothesis [decoupling of I-Xe chronometer from Ar-Ar chronometer has also been observed for chondrules from the EH3 chondrite Qingzhen, *Ash et al.* (1997)]. The best isochrons for three chondrules define closure ages of 1.3, 3.6 and 4.9 Ma after Bjurböle (Fig. 17). All of the chondrules display the regular I-Xe structure: the high temperature sites have higher values of $^{129}\text{I}/^{127}\text{I}$ than the low temperature sites, suggesting slow cooling or monotonic relaxation of the conditions during metamorphism (*Nichols et al.*, 1991). Using a non-diffusive, activation energy dependent model, cooling rates corresponding to a few hundred degrees per Ma, for the high temperature sites, down to a few degrees per Ma, for the low temperature sites, are estimated (*Nichols et al.*, 1991). This is the same range of values observed for the Allende CAIs and chondrules (*Swindle et al.*, 1983, 1988). These slow

"cooling" rates suggest that the post-formational processes in the regolith are likely responsible for the I-Xe fine structure.

Bridges et al. (1997) described a number of chondrules separated from Chainpur (LL3.4) and Parnallee (LL3.6) that contain mesostasis enriched in Na and Cl and contain microcrystalline sodalite, nepheline, and scapolite, and attributed these features to a pre-accretionary (which could be nebular or asteroidal) metasomatism. The I-Xe ages of the Parnallee chondrules (*Ash et al.*, 1995), which range from 1.75 ± 0.16 Ma to 5.0 ± 0.70 Ma after Bjurböle chondrule closure (Table 2), favor an asteroidal setting for the alteration. Two chondrules contain $^{128}\text{Xe}^*$, but no $^{129}\text{Xe}^*$, suggesting that they formed after the decay of ^{129}I , possibly by impact (*Ash et al.*, 1995). *Swindle et al.* (1991b) showed that the range of apparent I-Xe ages of Chainpur chondrules is ~ 50 Ma and that the chondrules evolved in a common reservoir with a chondritic I/Xe ratio. Based on these observations, *Swindle et al.* (199b) concluded that these ages reflect an asteroidal processing in a regolith.

The presence of halite (NaCl) and sylvite (KCl) containing inclusions of aqueous salt solutions in the H chondrite regolith breccias Monahans (1998) (H5) and Zag (H3-6) indicates that some of the aqueous alteration on the H chondrite parent body postdated thermal metamorphism (*Zolensky et al.*, 1999). We note, however, that there is no evidence that the halite in Zag and Monahans formed *in situ* (e.g., *Rubin et al.*, 2002). Based on the presence of secondary fluid inclusions in halite of both meteorites, *Zolensky et al.* (1999) concluded that aqueous activity occurred at low temperature ($< 50^\circ\text{C}$) and was episodic. A Rb-Sr model age for a halite crystal in Monahans (1998), calculated for an initial ratio of $^{87}\text{Sr}/^{86}\text{Sr} = 0.69876 \pm 0.00040$, the average for H-group chondrites, is 4.7 ± 0.2 Ga (*Zolensky et al.*, 1999). Subsequently, *Whitby et al.* (2000) reported essentially pure radiogenic ^{129}Xe in halite from Zag.

Correlated release of ^{129}Xe and ^{128}Xe corresponds to an initial ($^{129}\text{I}/^{127}\text{I}$) ratio of $(1.35\pm 0.05)\times 10^{-4}$ and an apparent formation time for the halite of 4.8 ± 0.9 Ma before the formation of Bjurböle reference chondrite, suggesting an early onset of aqueous activity on the Zag parent body. The retention of a high $^{129}\text{Xe}^*/^{127}\text{I}$ ratio implies that halite has not been subjected to substantial dissolution and recrystallization in the 4.5 Ga since its formation, suggesting that the processes that led to aqueous activity on the Zag parent body may have ended quickly after evaporation of water into space (*Whitby et al.*, 2000).

3.7. Timescale of Alteration of Enstatite Chondrites

Ash et al. (1997) reported apparent I-Xe ages of chondrules from the EH3 enstatite chondrite Qingzhen. Most chondrules give excellent isochrons with errors < 1 Ma; only one of the chondrules shows evidence for a slight isotopic disturbance. The observed range in I-Xe ages, from -1.08 to +1.98 relative to Shallowater (Table 2), is comparable to those in most unequilibrated ordinary chondrites. Based on the apparent lack of evidence for secondary alteration in Qingzhen, these ages were interpreted as primary, corresponding to the ages of chondrule formation. The iodine carrier in enstatite chondrites is unknown, but the presence of sodalite-like mesostases in some type I chondrules in Qingzhen suggests that it could be sodalite. The origin of these mesostases, and the interpretation of I-Xe ages, remain unclear.

4. SUMMARY AND FUTURE WORK

Mineralogical, petrographic, and isotopic observations indicate that most groups of chondritic meteorites experienced asteroidal alteration to various degrees, resulting in formation of secondary minerals such as phyllosilicates, magnetite, carbonates, ferrous olivine (Fa_{40-100}), salite-hedenbergite pyroxenes ($\text{Fs}_{10-50}\text{Wo}_{45-50}$), wollastonite, andradite, nepheline, pentlandite,

pyrrhotite, Fe,Ni-carbides, and Ni-rich metal. The alteration occurred in the presence of aqueous solutions under variable conditions (temperature, water/rock ratio, fO_2 , and fluid compositions) and in many cases was multistage. Although some alteration predated agglomeration of the final chondrite asteroidal bodies (e.g., dark inclusions in CV chondrites), there is no compelling evidence that the alteration occurred in the solar nebula nor in planetesimals of earlier generations. The ^{26}Al - ^{26}Mg , ^{53}Mn - ^{53}Cr , and ^{129}I - ^{129}Xe dating of secondary minerals suggests that alteration may have started within 1-2 Ma after formation of the CV CAIs having absolute Pb-Pb age of 4567.2 ± 0.6 Ma and lasted up to 15 Ma (Tables 1-3; Figs. 18, 19). Based on these observations, we infer that the chondrite parent bodies must have accreted within the first 1-2 Ma after collapse of the protosolar molecular cloud.

There are several carbonaceous chondrite groups not discussed in this chapter with clear evidence for secondary alteration; these include CR, CH, CB, and CO chondrites. The CR chondrites experienced aqueous alteration to various degrees that resulted in formation of phyllosilicates, magnetite, and carbonates (e.g., *Krot et al.*, 2002). The CB and CH chondrites contain heavily aqueously-altered clasts composed of phyllosilicates, framboidal and platelet magnetite, and carbonates (*Greshake et al.*, 2002). The CO chondrites experienced alteration similar to that observed in CV chondrites (*see CV chondrites and their alteration*). The alteration resulted in formation of nepheline, sodalite, ferrous olivine, magnetite, Fe,Ni-carbides, and Ni-bearing sulfides (e.g., *Jones*, 1997a,b; *Rubin*, 1998; *Russell et al.*, 1998; *Chizmadia et al.*, 2002; *Itoh and Tomeoka*, 2003). A degree of alteration correlates with petrologic types of the host meteorites, suggesting that it occurred in an asteroidal setting (e.g., *Itoh and Tomeoka*, 2003). Although the secondary mineralization in the CR, CO, CB, and CH chondrites has been well-documented, there have yet been no attempts made to date it.

Future studies of isotope dating of secondary mineralization of chondritic meteorites should also be focused on understanding the multistage alteration histories using combinations of analytical tools, including SEM, EPMA, CL, TEM, SIMS, and ICP-MS. This approach has already been successfully used in dating carbonate formation in CM carbonaceous chondrites (*Brearley et al.*, 1999, 2001; *Brearley and Hutcheon*, 2000, 2002). Small grain sizes of the secondary minerals suitable for *in situ* Mn-Cr isotope dating (e.g., carbonates, ferrous olivine) will probably require use of NanoSIMS (e.g., *Hoppe et al.*, 2004).

Finally, we would like to emphasize that progress in the chronology of the early solar system processes requires better understanding the origin of short-lived radionuclides [external (injection) *vs.* internal (irradiation)] and their distribution (homogeneous *vs.* heterogeneous) in the protoplanetary disk (e.g., *Goswami et al.*, 2000, 2004; *Gounelle et al.*, 2001), and establishing a unified chronology of the early solar system processes using these radionuclides (e.g., *Gilmour and Saxton*, 2001; *Gilmour et al.*, 2004). These issues remain unresolved.

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Table 1. The I-Xe ages of the dark inclusions from the reduced and oxidized CV chondrites relative to the age of Shallowater (4563.5 ± 1.0 Ma; *Gilmour et al.*, work in progress) internal standard.

chondrite/classif.	sample	I-Xe age, Ma	ref.
Allende CV _{oxA}	1a-1	-2.8 ± 0.3	[1]
	12b-1	-2.0 ± 0.3	
	4294-1	-1.9 ± 0.3	
	4a1/b1	-1.9 ± 0.3	
	IV-1	-1.9 ± 0.2	
	14b-1	-1.6 ± 0.2	
	4884-2	-1.5 ± 0.2	
	4301-1	-1.5 ± 0.1	
	4884-1	-1.4 ± 0.8	
	4314-3	-1.1 ± 0.2	
	1-3	-1.1 ± 0.2	
	25sl-tw1	-1.1 ± 0.2	
	4320-1	-1.0 ± 0.3	
	4884-6	-1.0 ± 0.2	
	IV-2	-0.8 ± 0.3	
	4884-5	-0.7 ± 0.2	
4884-3	-0.5 ± 0.3		
Efremovka CV _{red}	E53	-4.9 ± 1.8	[2, 3]
	E39	$+0.8 \pm 2.0$	
	E80	-1.0 ± 0.5	
Leoville CV _{red}	LV1	$+3.0 \pm 0.1$	[4]
	LV2	$+9.5 \pm 2.3$	
Vigarano CV _{breccia}	2226	$+8.8 \pm 0.6$	

References: [1] *Pravdivtseva et al.* (2003b); [2] *Krot et al.* (1999); [3] *Swindle et al.* (1998); [4] *Pravdivtseva et al.* (2003c).

Table 2. The I-Xe ages of chondrules from type 3 ordinary and enstatite chondrites relative to the age of Shallowater (4563.5 ± 1.0 Ma; *Gilmour et al.*, work in progress) internal standard.

chondrite/classif.	chd#	I-Xe age, Ma	ref.
Semarkona, LL3.0	CD-159(l)	$+4.9 \pm 0.5$	[1]
	CD-159(h)	-4.9 ± 2.9	
	CD-92	-1.9 ± 1.1	
	CD-95	$+4.1 \pm 1.2$	
	CD-54	-0.4 ± 1.2	
	CD-79	$+0.6 \pm 1.7$	
	CD-60	-2.4 ± 1.7	
	CD-173	-1.8 ± 2.1	
	CD-160	$+4.7 \pm 1.1$	
	CD-8(l)	$+0.6 \pm 1.1$	
	CD-8(h)	-4.5 ± 2.3	
	CD-84(l)	$+0.8 \pm 0.3$	
	CD-84(h)	-2.5 ± 0.9	
	CD-129	$+1.3 \pm 1.0$	
	CD-153	-1.8 ± 2.8	
	CD-169	0 ± 0.3	
	CD-139	-2.6 ± 2.5	
	CD-101	-4.2 ± 0.6	
CD-174	-1.9 ± 1.7		
Parnallee, LL3.4	CB1	$+4.16 \pm 0.44$	[2]
	CB2	$+1.29 \pm 0.16$	
	P6	$+4.54 \pm 0.70$	
	P9	no $^{129}\text{Xe}^*$	
	Feline	$+5.05$	
	P32	$+1.94 \pm 0.26$	
	MC1	no $^{129}\text{Xe}^*$	
Qingzhen, EH3	QC1	$+1.98$	[3]
	QC3	$+0.44$	
	QC4	-1.08	
	QC5	$+1.41$	
	QC6	$+0.10$	
	QC7	$+0.64$	
	QC8	$+1.70$	

References: [1] *Swindle et al.* (1991a); [2] *Ash et al.* (1995); [3] *Ash et al.* (1997).

Table 3. The initial $^{53}\text{Mn}/^{55}\text{Mn}$ ratios in secondary carbonate and fayalite in carbonaceous chondrites and their Mn-Cr ages relative angrite LEW86010 (4557.8 ± 0.5 Ma).

chondrite	classification	mineral analyzed	$(^{53}\text{Mn}/^{55}\text{Cr})_0$	age relative LEW86010	ref.
Orqueil & Ivuna	CI	dolomite	$(1.99 \pm 0.16) \times 10^{-6}$	-2.5 ± 0.7	[1]
Orqueil	CI	dolomite	$(1.42 \pm 0.16) \times 10^{-6}$	-0.7 ± 0.9	[1]
Orqueil	CI	breunnerite	$(1.97 \pm 0.18) \times 10^{-6}$	-2.5 ± 0.8	[2]
Orqueil	CI	breunnerite	$(3.4 \pm 0.4) \times 10^{-6}$	-5.3 ± 0.9	[2]
Orqueil	CI	Cr-carbonates	3.4×10^{-6}	-5.3	[3]
Supuhee, clast	CI-like	carbonates calcite,	$(8 \pm 4) \times 10^{-6}$	-9.9 ± 2.5	[3]
Kaidun*	breccia	dolomite	$(9.4 \pm 1.6) \times 10^{-6}$	-10.8 ± 1.1	[4]
ALH84034	CM1	dolomite	$(5.0 \pm 1.5) \times 10^{-6}$	-7.4 ± 1.7	[5]
Y791198	CM2	calcite	$(8.7 \pm 1.5) \times 10^{-6}$	-10.3 ± 2.2	[6]
Kaba	CV _{oxB}	fayalite	$(2.32 \pm 0.18) \times 10^{-6}$	-3.3 ± 0.7	[7]
Mokoia	CV _{oxB}	fayalite	$(2.28 \pm 0.37) \times 10^{-6}$	-3.2 ± 1.0	[8]
MAC88107	ungrouped	fayalite	$(1.58 \pm 0.28) \times 10^{-6}$	-1.3 ± 1.2	[9]

References: 1 - *Endress et al.* (1996); 2 - *Hutcheon and Phinney* (1996); 3 - *Hutcheon et al.* (1997); 4 - *Hutcheon et al.* (1999); 5 - *Brearley and Hutcheon* (2000); 6 - *Brearley et al.* (2001); 7 - *Hutcheon et al.* (1998); 8 - *Hua et al.* (2002); 9 - *Krot et al.* (2000a). *contains CR-, C1- and CM-like materials.

FIGURE CAPTIONS

Fig. 1. ^{53}Mn - ^{53}Cr evolution diagrams for carbonates in CI chondrites. a - Dolomite fragments from Orgueil and Ivuna; b - an expanded-scale view of the lower left-hand corner of this plot. The labels a, b and c denote different spots on a given dolomite fragment. The line of slope 1.99×10^{-6} is a best-fit line through all data points for Orgueil 5 and normal Cr (that is, $\delta^{53}\text{Cr} = 0$ at $^{55}\text{Mn}/^{52}\text{Cr} = 0$). The data points for Orgueil 8 and Ivuna 2 fragments are consistent with this line, the data points for the remaining two fragments, Orgueil 1 and 2, fall close to the line of slope 1.42×10^{-6} but, compared to the analytical errors, the deviations are not large enough to clearly establish that different carbonates formed at different times (from *Endress et al.*, 1996). c, d - Breunnerite and dolomite in Orgueil (c) and dolomite in Supuhee (d) CI carbonaceous chondrites. Different symbols represent different grains. The lines of slope $(1.97 \pm 0.18) \times 10^{-6}$ and $(8 \pm 4) \times 10^{-6}$ are the best-fit lines through all data points and normal Cr; error bars are 2σ (from *Hutcheon and Phinney*, 1996; *Hutcheon et al.*, 1997).

Fig. 2. ^{129}I - ^{129}Xe evolution diagrams for the Orgueil magnetic separates and Shallowater aubrite. The ^{129}I - ^{129}Xe ages for the nearly pure magnetite fraction containing >90% magnetite predate Shallowater by 1.9 ± 0.2 Ma. For the highly magnetic fraction composed of ~14% magnetite and ~86% hydrated carbonaceous material, the I-Xe age is 3.0 ± 0.4 Myr Ma younger than Shallowater (4563.5 ± 1 Ma; *Gilmour et al.*, work in progress), suggesting that the magnetic fraction may contain several iodine carriers recording different stages of aqueous activity on the CI parent body; error bars are 1σ (data from *Hohenberg et al.*, 2000 and *Pravdivtseva et al.*, 2003b).

Fig. 3. Oxygen isotopic compositions of separated components from CI and CM chondrites. Phyllosilicate-rich matrix (phyl) + carbonates and magnetite (mgt) in CI chondrites, as well as

phyllosilicate-rich matrix and carbonates in CM chondrites are out of isotopic equilibrium. Similar $\Delta^{17}\text{O}$ values for calcite and dolomite fractions from the same splits of the same CM chondrites indicate that both minerals in each split precipitated from a single fluid reservoir. The terrestrial fractionation (TF) line is shown for reference (data from *Rowe et al.*, 1994; *Leshin et al.*, 2001; *Benedix et al.*, 2003).

Fig. 4. ^{53}Mn - ^{53}Cr evolution diagrams for carbonates from (a) Kaidun, and CM carbonaceous chondrites (b) Y791198 and (c) ALH84034. Different symbols represent different grains. The lines of slope $(9.4\pm 1.6)\times 10^{-6}$, $(8.7\pm 1.5)\times 10^{-6}$, and $(5.0\pm 1.5)\times 10^{-6}$ are the best-fit lines through all data points and normal Cr; error bars are 2σ (data from *Hutcheon et al.*, 1999; *Brearley et al.*, 2001; *Brearley and Hutcheon*, 2000).

Fig. 5. Backscattered electron (BSE) images of (a, b) a porphyritic olivine-pyroxene type I chondrule, (c) isolated fayalitic olivine grain and (d) magnetite-sulfide nodule in the ungrouped carbonaceous chondrite MAC88107. a, b - The chondrule is surrounded by a continuous fine-grained rim (FGR) crosscut by fayalite (fa) - hedenbergite (hed) - magnetite (mgt) veins. The veins start at the opaque nodules composed of Ni-bearing sulfide and magnetite in the peripheral portion of the chondrule. Chondrule mesostasis (lm) is largely leached out, whereas forsteritic olivine (fo) and low-Ca pyroxene (px) phenocrysts appear to be unaltered. c - Fayalitic olivine (fa ol) is overgrown by fayalite. d - Fayalite preferentially replaces magnetite of the sulfide-magnetite nodule. Low-Ca pyroxene grains at the contact with fayalite and magnetite and forsteritic olivine grain overgrown by fayalite appear to be unaltered (after *Krot et al.*, 2000a).

Fig. 6. ^{53}Mn - ^{53}Cr evolution diagrams for (a) a fayalite grain from the ungrouped carbonaceous chondrite MAC88107, (b) four fayalite grains in three porphyritic olivine-pyroxene type I chondrules from the CV_{oxB} chondrite Mokoia, and (c) twelve fayalite grains in matrix of the

CV_{oxB} chondrite. The lines of slope $(1.58 \pm 0.26) \times 10^{-6}$, $(2.32 \pm 0.18) \times 10^{-6}$ and $(2.28 \pm 0.37) \times 10^{-6}$, are the best-fit lines through the data points and normal Cr; error bars are 2σ (data from *Hutcheon et al.*, 1998; *Krot et al.*, 2000a; *Hua et al.*, 2002).

Fig. 7. BSE images of matrices in the oxidized CV chondrites (a-b) and Allende dark inclusions (d). All matrices contain Ca,Fe-pyroxenes-andradite (hed-andr) nodules. Matrix in the CV_{oxB} Kaba contains nearly pure fayalite (\sim Fa₁₀₀) and very fine-grained groundmass largely composed of ferrous olivine (\sim Fa₅₀) and phyllosilicates. Matrix in the CV_{oxA-B} MET00430 contains fayalite grains showing inverse compositional zoning (Fa₈₀₋₅₀) and coarser grained lath-shaped ferrous olivine (\sim Fa₅₀). Matrices in the CV_{oxA} ALH81258 and Allende dark inclusion contain relatively coarse-grained, lath-shaped compositionally uniform (\sim Fa₅₀) ferrous olivine.

Fig. 8. BSE images of different textural occurrences of secondary fayalite in the CV_{oxB} chondrites Kaba and Mokoia. a, b - Porphyritic olivine-pyroxene (POP) type I chondrule surrounded by a continuous fine-grained rim crosscut by fayalite (fa) - magnetite (mgt) veins. The veins start at the opaque nodules composed of Ni-bearing sulfide (sf) and magnetite in the peripheral portion of the chondrule. Region outlined in "a" is shown in detail in "b". c - Opaque nodule in type I chondrule replaced by magnetite, Ni-bearing sulfides, fayalite, and salite-hedenbergite pyroxenes (hed). d - Opaque nodule within type I POP chondrule; numbers correspond to fayalite content (in mol%). Magnetite is replaced by pure fayalite (Fa₁₀₀); forsterite phenocrysts (Fa₁) are partly pseudomorphed by ferrous olivine (Fa₆₃); an outline of one of the grains is indicated by arrows. Fayalite is crosscut by a vein of ferrous olivine (Fa₈₇), suggesting that forsterite is the source of Mg. e - Amoeboid olivine aggregate composed of forsterite, spinel, Al-diopside, and anorthite. Forsterite grains are overgrown by euhedral ferrous olivines ranging in compositions from Fa_{<50} to Fa₇₃; some of the fayalite grains contain inclusions of Fe,Ni-

sulfides (sf). f - Fine-grained CAI consisting of concentrically-zoned objects composed of spinel (sp) surrounded by phyllosilicates (phyl) and Al-diopside (di); phyllosilicates probably replace primary anorthite or melilite. Euhedral fayalite grains occur between these bodies; Ca,Fe-pyroxenes (hed) overgrow Al-diopside.

Fig. 9. Oxygen isotopic compositions of secondary magnetite (Mgt), fayalite (Fa), Ca,Fe-rich pyroxenes (CaFe-px), andradite (Andr), and wollastonite (Wol), and primary forsteritic olivine (Fo) (a) in type I chondrules in the CV_{oxB} chondrites Kaba and Mokoia (data from *Choi et al.*, 2000; *Hua et al.*, 2003), (b) in chondrules, matrix (mx), and in rims around CAIs (data from *Choi et al.*, 2000; *Cosarinsky et al.*, 2003), and (c) in and around Allende dark inclusions (data from *Krot et al.*, 2000c); error bars are 2σ . The terrestrial fractionation (TF) line and carbonaceous chondrite anhydrous mineral (CCAM) line are shown for reference. (a) In Mokoia, the magnetite and fayalite differ in $\delta^{18}\text{O}$ by $\sim 20\text{‰}$, suggesting formation at low-temperature. In Kaba, the compositions of fayalite and magnetite reported by *Choi et al.* (2000) are nearly identical, and very close to the intersection of the TF and CCAM lines. The compositions of Kaba fayalites reported by *Choi et al.* (2000) are inconsistent with those reported by *Hua et al.* (2003); the latter are similar to those of Mokoia fayalites. We note that compositions of fayalite and magnetite in Kaba reported by *Choi et al.*, 2000) were collected with a 3 month interval and might be in error. Compositions of forsteritic olivine phenocrysts plot along CCAM line and are not in equilibrium with those of the secondary minerals. (b) Oxygen isotope compositions of Ca,Fe-rich pyroxenes and andradite in matrix (mx) and in rims around CAIs are similar and plot parallel to the TF with a range in $\delta^{18}\text{O}$ of $\sim 20\text{‰}$, suggesting formation at low-temperature. Oxygen isotope compositions of magnetite overlap with those of Ca,Fe-pyroxenes and andradite, but plot largely to the left from CCAM line.

Fig. 10. ^{129}I - ^{129}Xe evolution diagrams for mineral fractions separated from the CV_{oxB} chondrites (a-c) Kaba and (d-f) Bali; error bars are 1σ . The I-Xe ages shown are relative to the Shallowater internal standard (4563.5 ± 1 Ma; *Gilmour et al.*, work in progress). Numbers next to points represent extraction W-coil temperatures in $^{\circ}\text{C}$ (the sample is probably $150\text{-}200^{\circ}\text{C}$ cooler).

Fig. 11. a-d - BSE images of secondary minerals in POP type I chondrules in the CV_{oxA} chondrite ALH84028. Magnetite-sulfide nodules are replaced by ferrous olivine (fa) and Ca,Fe-pyroxenes (CaFe-px); both contain abundant inclusions of sulfides (sf). Low-Ca pyroxene phenocrysts (px) are replaced by ferrous olivine. Chondrule mesostasis is replaced by nepheline (nph). Forsteritic olivine (fo) phenocrysts are largely unaltered, but show enrichment in fayalite contents near the edges and along the fractures. e, f - BSE images of secondary fayalite in the $\text{CV}_{\text{oxA-B}}$ chondrite MET00430. e - Fayalite overgrowing olivine-pyroxene chondrule fragment shows inverse compositional zoning (Fa_{75-50}). f - Euhedral fayalite grain overgrowing low-Ca pyroxene (px) phenocryst in outer part of a type I chondrule shows complex chemical zoning suggesting dissolution of fayalite and precipitation of more forsteritic olivine from a fluid phase. Numbers correspond to fayalite contents (from *Krot et al.*, 2004a).

Fig. 12. a-c - ^{129}I - ^{129}Xe evolution diagrams for fine-grained CAIs in Allende (from *Pravdivtseva et al.*, 2003b). The contribution from trapped Xe component is within experimental uncertainty consistent with the "planetary" OC-Xe (*Lavielle and Marti*, 1992). d-f - ^{129}I - ^{129}Xe evolution diagrams for Allende dark inclusions. Two isochrons plotted for the dark inclusion IV-1 correspond to low- and high-temperature Xe released. All isochrons suggest "sub-planetary" trapped components (*Hohenberg et al.*, 2004). Error bars are 1σ . I-Xe ages are relative to the Shallowater internal standard (4563.5 ± 1 Ma; *Gilmour et al.*, work in progress).

Fig. 13. BSE images of altered chondrules in the dark inclusions (DI) 3529 (a), 4301-2 (b), E53 (c, d), and E39 (e, f) in the oxidized CV chondrite Allende (a, b) and in the reduced CV chondrite Efremovka (c-f). a, b - Chondrules are replaced to various degrees by ferrous olivine, nepheline (nph; black in "d"), and Ca,Fe-pyroxenes (Ca,Fe-px). Chondrule shown in "a" is surrounded by a fine-grained rim composed of lath-shaped ferrous olivine (fa) and nepheline. The rim is crosscut by a vein composed of Ca,Fe-pyroxenes and Fe,Ni-sulfides (white). The vein starts at the opaque nodule (outlined) that is replaced by Ca,Fe-pyroxenes and ferrous olivine; sulfide grains (white) are relict. c, d - Chondrules in E53 are pseudomorphed to a various degree by a fine-grained mixture of ferrous olivine (fa) and very minor phyllosilicates (phyl). e, f - Chondrules in E39 are nearly completely replaced by a fine-grained mixture of ferrous olivine, phyllosilicates, and andradite (andr). Forsteritic olivine (fo) and high-Ca pyroxenes (cpx) are relict. mes = mesostasis; met = Fe,Ni-metal (from *Krot et al.*, 1998a, 1999).

Fig. 14. Ca $K\alpha$ X-ray elemental maps of the heavily-altered dark inclusions 4301-2 (a) and IV-1 (b) in the oxidized CV chondrite Allende. The dark inclusions (DI) contain chondrule pseudomorphs (indicated by stars) which are depleted in Ca and consist of the secondary ferrous olivine, nepheline, sodalite, and Fe,Ni-sulfides (see Fig. 14b). The dark inclusion 4301-2 is crosscut by multiple veins composed of Ca,Fe-pyroxenes and andradite. Both dark inclusions are surrounded by continuous Ca-rich rims composed of Ca,Fe-pyroxenes, andradite, wollastonite, and kirschsteinite. The outer portions of the dark inclusions are depleted in Ca, whereas the neighboring matrix of Allende contains abundant Ca,Fe-rich nodules composed of Ca,Fe-pyroxenes, andradite, and wollastonite, suggesting that Ca lost from the dark inclusions precipitated as rims and nodules around them (from *Krot et al.*, 2001).

Fig. 15. ^{129}I - ^{129}Xe evolution diagrams for the Efremovka dark inclusions E53, E39 (data from Swindle *et al.*, 1998; Krot *et al.*, 1999), and E80; error bars are 1σ (data from Pravdivtseva *et al.*, 2003c). The ages shown are relative to the Shallowater internal standard (4563.5 ± 1 Ma; Gilmour *et al.*, work in progress). The two apparent isochrons for E80 correspond to different peaks in the release profiles of radiogenic ^{128}Xe and ^{129}Xe , suggesting that E80 contains two different iodine-carrying mineral phases with the same closure time but different trapped components. The circled temperature points represent intermediate extraction steps between these two release peaks where radiogenic ^{128}Xe and ^{129}Xe do not correlate.

Fig. 16. Representative three-isotope plots for Xe from irradiated Semarkona chondrules. These include one sample with dual isochrons (CD-159), and samples with single isochrons with apparent old (CD-92 and CD-101) and young (CD-160) I-Xe ages. Diamonds denote points included in high-temperature isochrons and circles are those included in low-temperature (or single) isochrons; errors bars are 1σ . Numbers next to points represent extraction coil temperatures in $^{\circ}\text{C}$ (the sample is probably 200-300 $^{\circ}\text{C}$ cooler) (from Swindle *et al.*, 1991a).

Fig. 17. ^{129}I - ^{129}Xe evolution diagrams for Tieschitz chondrules. Numbers next to points represent extraction coil temperatures in $^{\circ}\text{C}$ (the sample is probably 200-300 $^{\circ}\text{C}$ cooler); errors bars are 2σ (from Nichols *et al.*, 1991).

Fig. 18. Mn-Cr ages of the secondary carbonates and fayalite in carbonaceous chondrites relative to the LEW86010 angrite; errors are 2σ (data from Endress *et al.*, 1996; Hutcheon and Phinney, 1996; Hutcheon *et al.*, 1997, 1998, 1999; Brearley and Hutcheon, 2000); Brearley *et al.*, 2001; Hua *et al.*, 2002; Krot *et al.*, 2000a). Absolute ages of CAIs from CV chondrites (4567.2 ± 0.6 Myr; Amelin *et al.*, 2002) and ages calculated based on the initial $^{53}\text{Mn}/^{55}\text{Mn}$ ratios of 1.4×10^{-5}

(Lugmair and Shukolyukov, 2001), $(2.8 \pm 0.3) \times 10^{-5}$ (Nyquist *et al.*, 2001), and 4.4×10^{-5} (Birck and Allègre, 1988; Birck *et al.*, 1999) are plotted for reference.

Fig. 19. I-Xe ages of the CV chondritic components (CAIs, chondrules, matrix, dark inclusions) and mineral fractions (magnetite, phyllosilicates) relative to the Shallowater aubrite internal standard; errors are 1σ (data from Swindle *et al.*, 1983, 1988, 1998; Krot *et al.*, 1999; Hohenberg *et al.*, 2001; Pravdivtseva *et al.*, 2003b,c). Based on the comparison of I-Xe and Mn-Cr systems with the absolute Pb-Pb chronometer for samples analysed by multiple isotope systems, Gilmour *et al.* (work in progress) infer that the I-Xe system closed in Shallowater aubrite at 4563.5 ± 1.0 Ma before the present, i.e. 5.7 ± 1.1 Ma earlier than the Mn-Cr system closed in LEW86010 angrite.

Fig. 1.

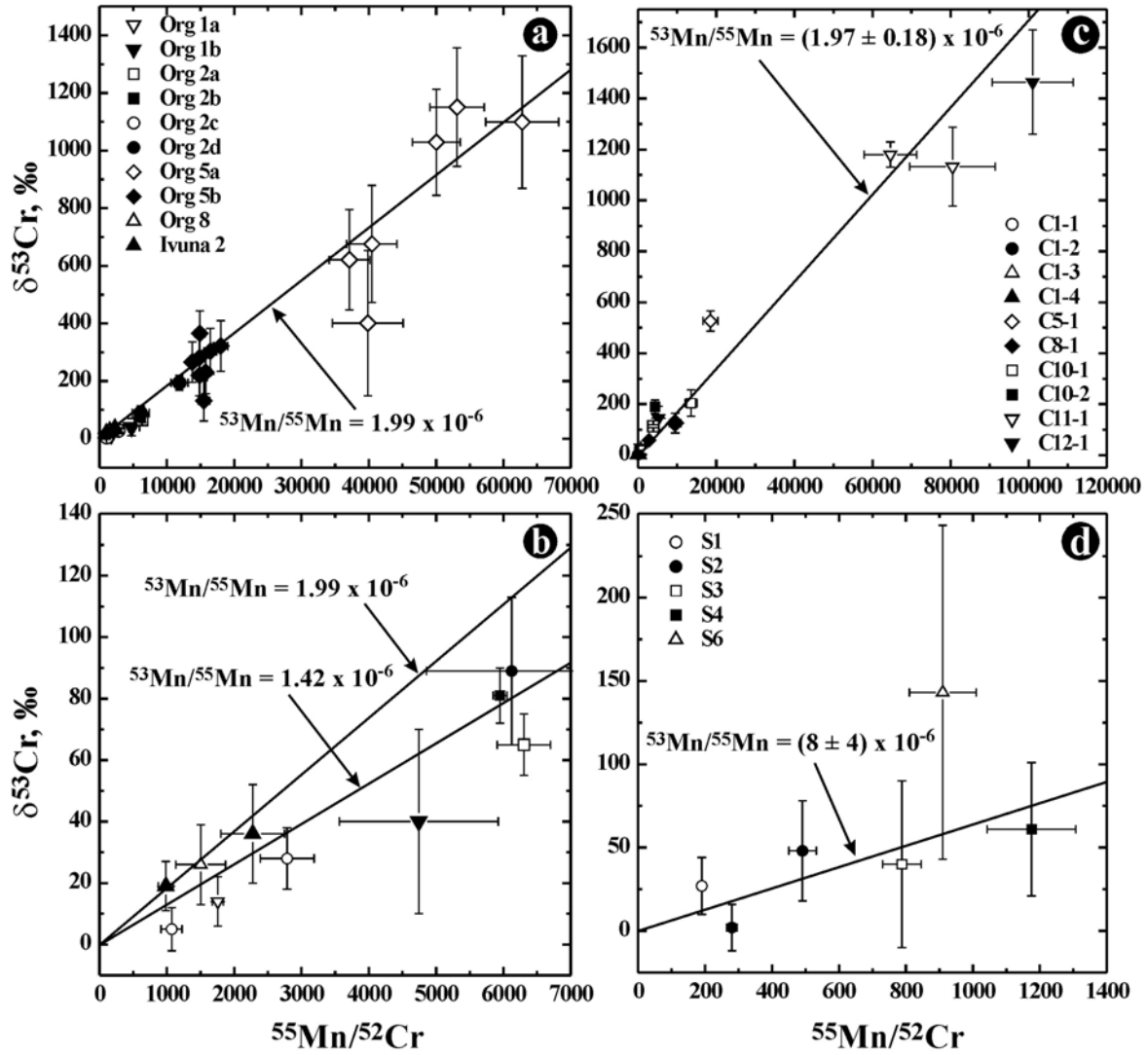


Fig. 2.

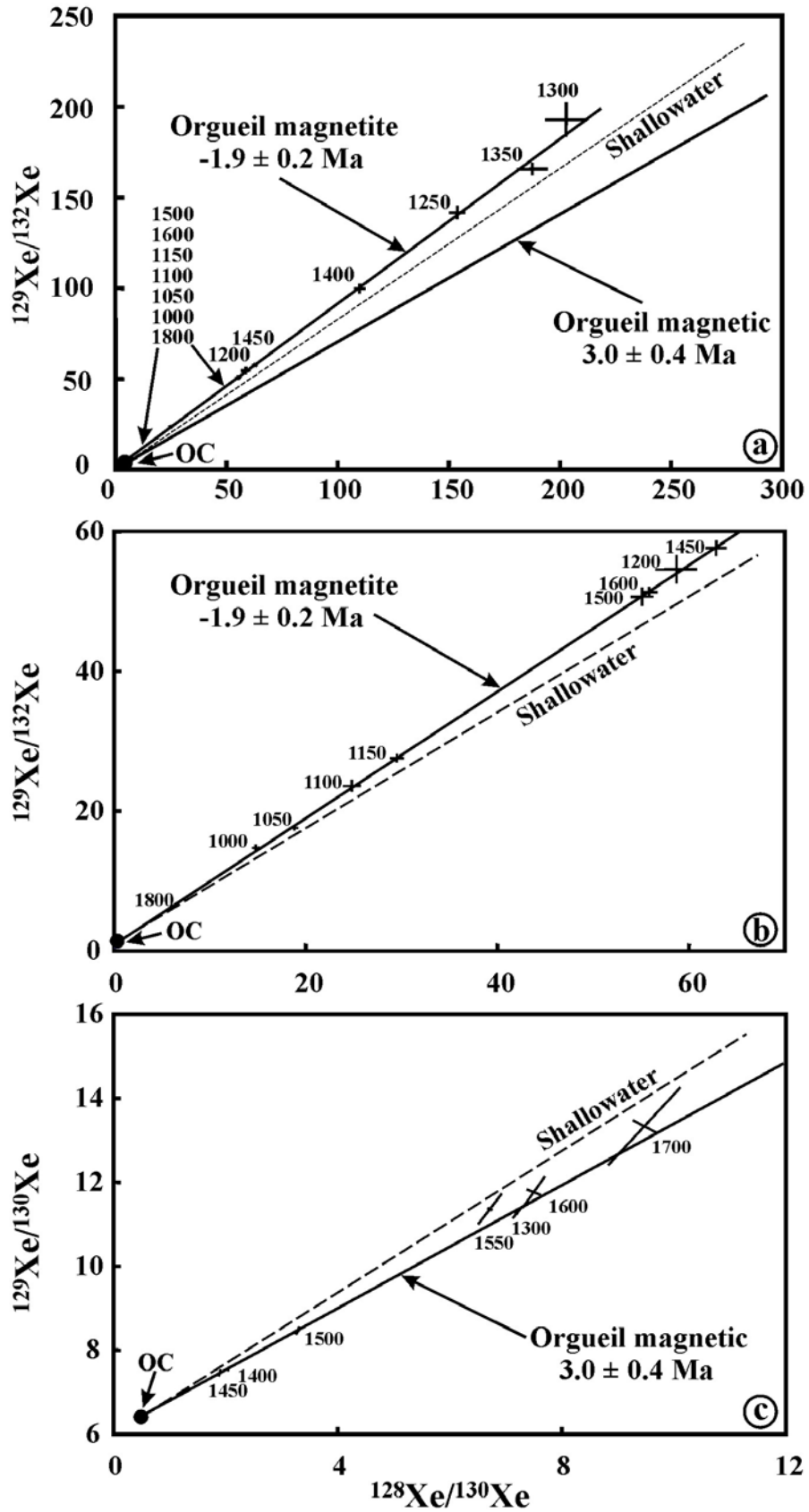


Fig. 3.

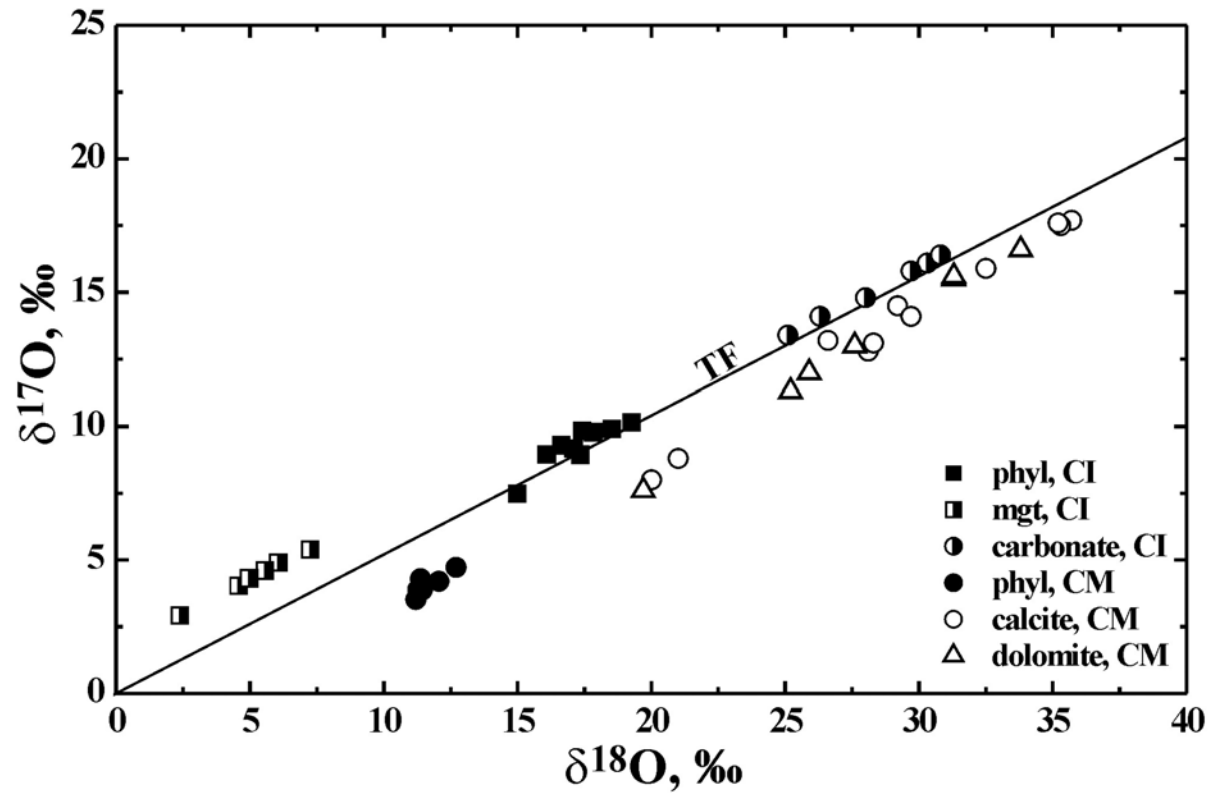


Fig. 4.

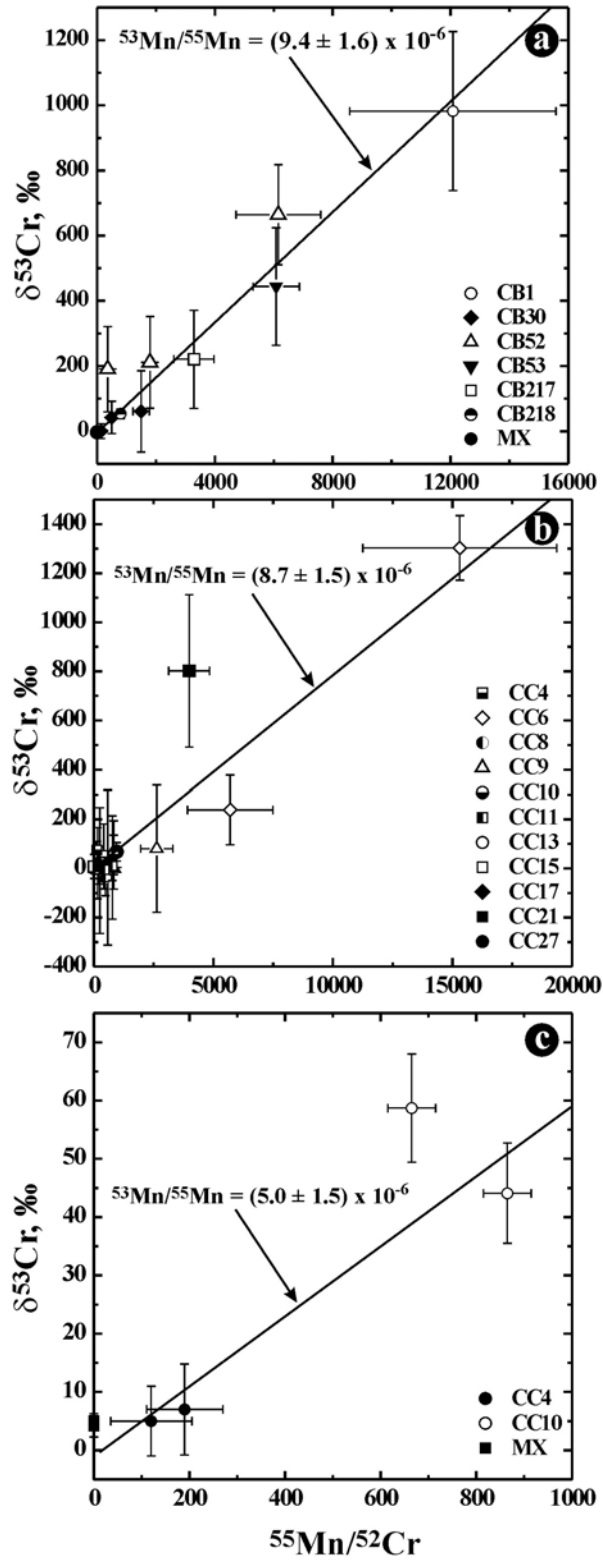


Fig. 5.

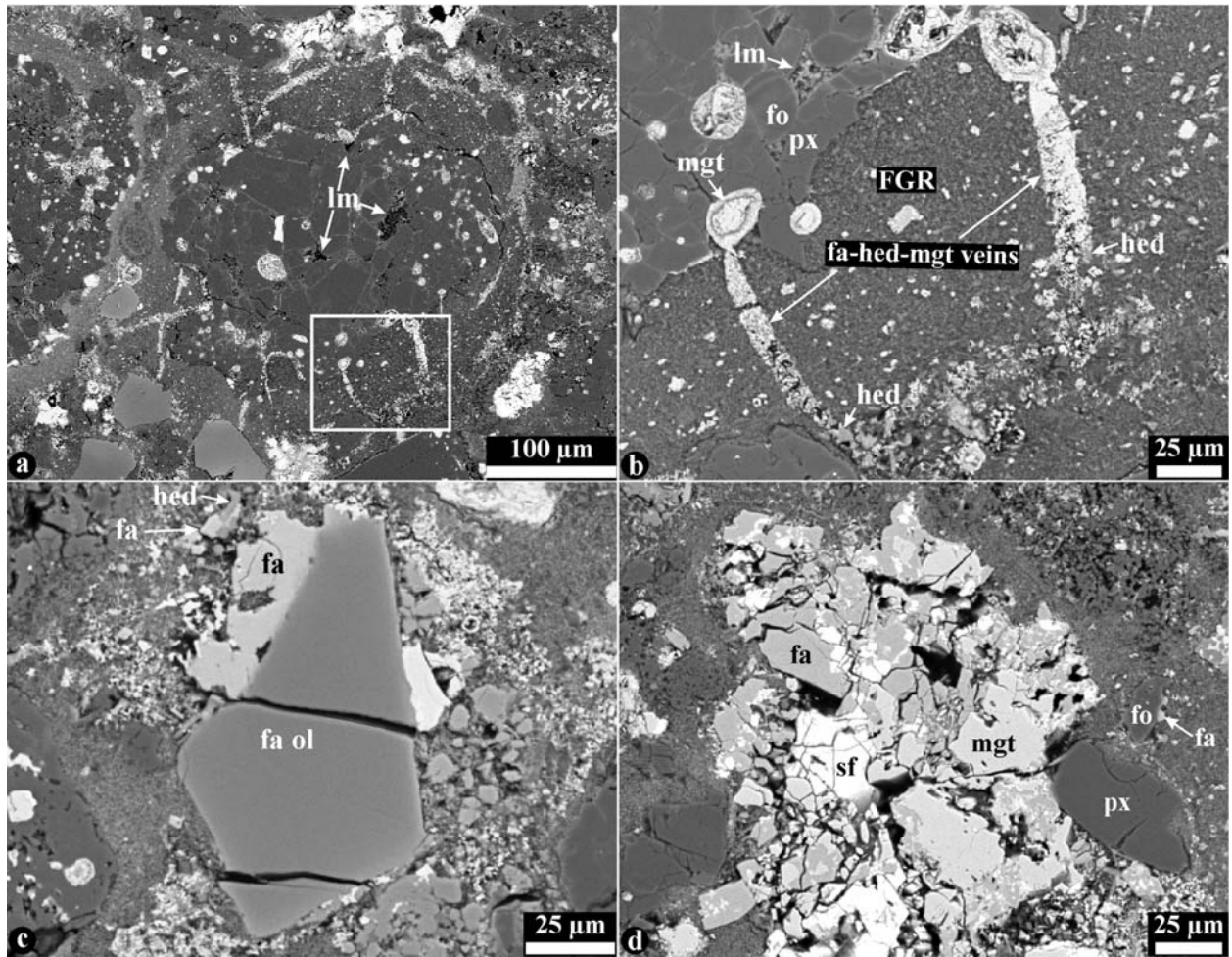


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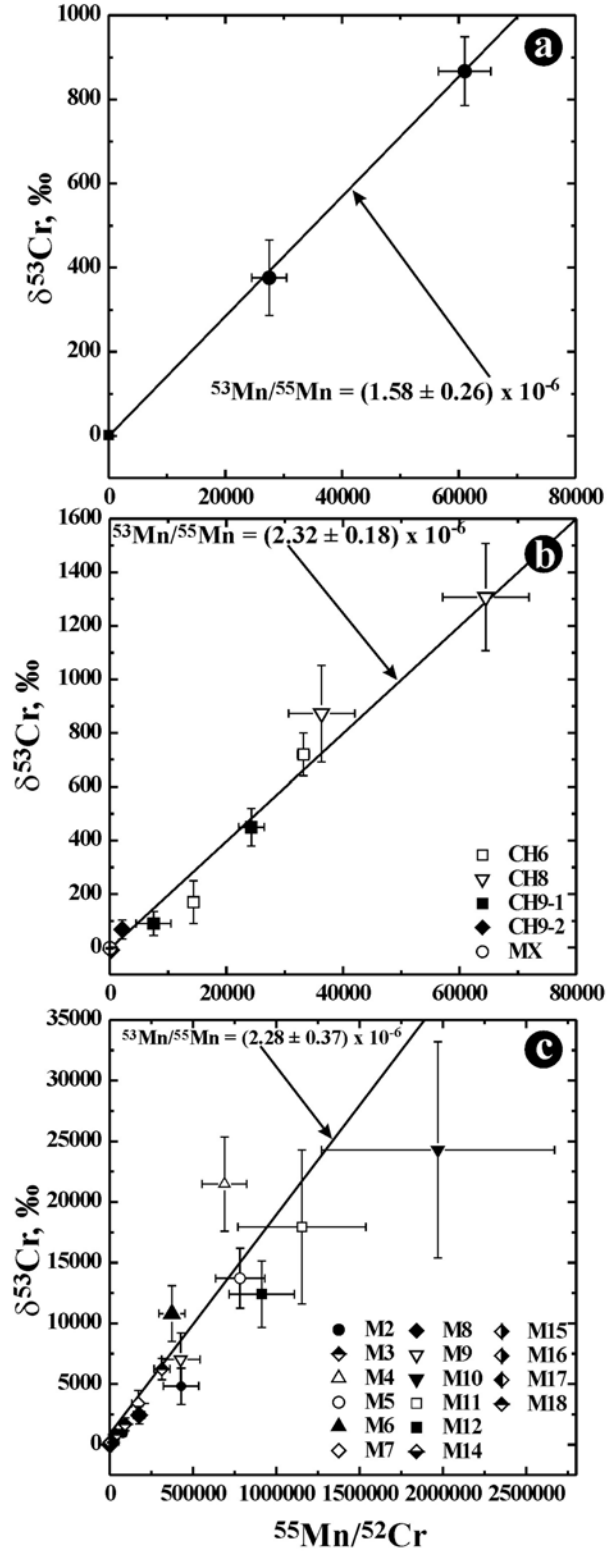


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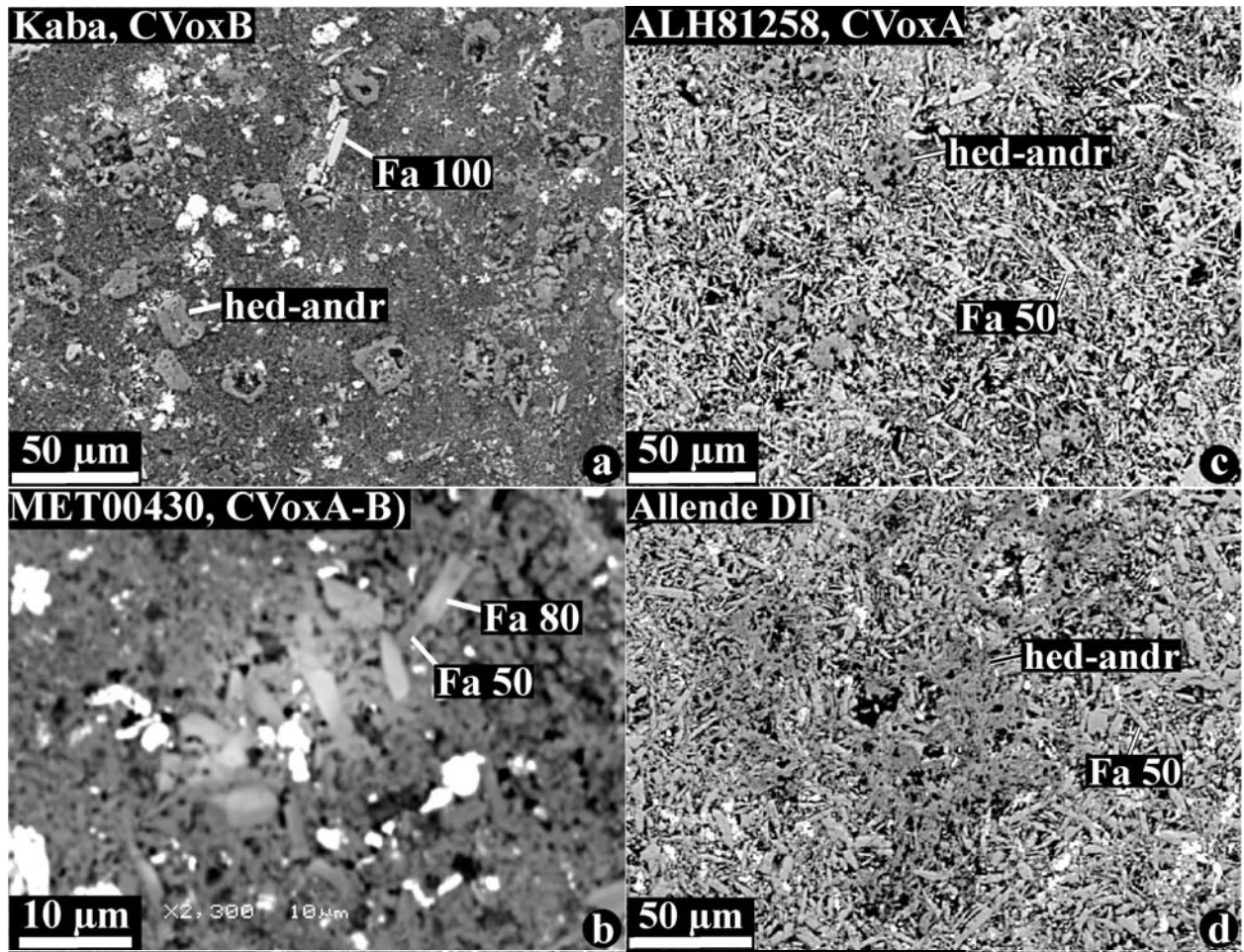


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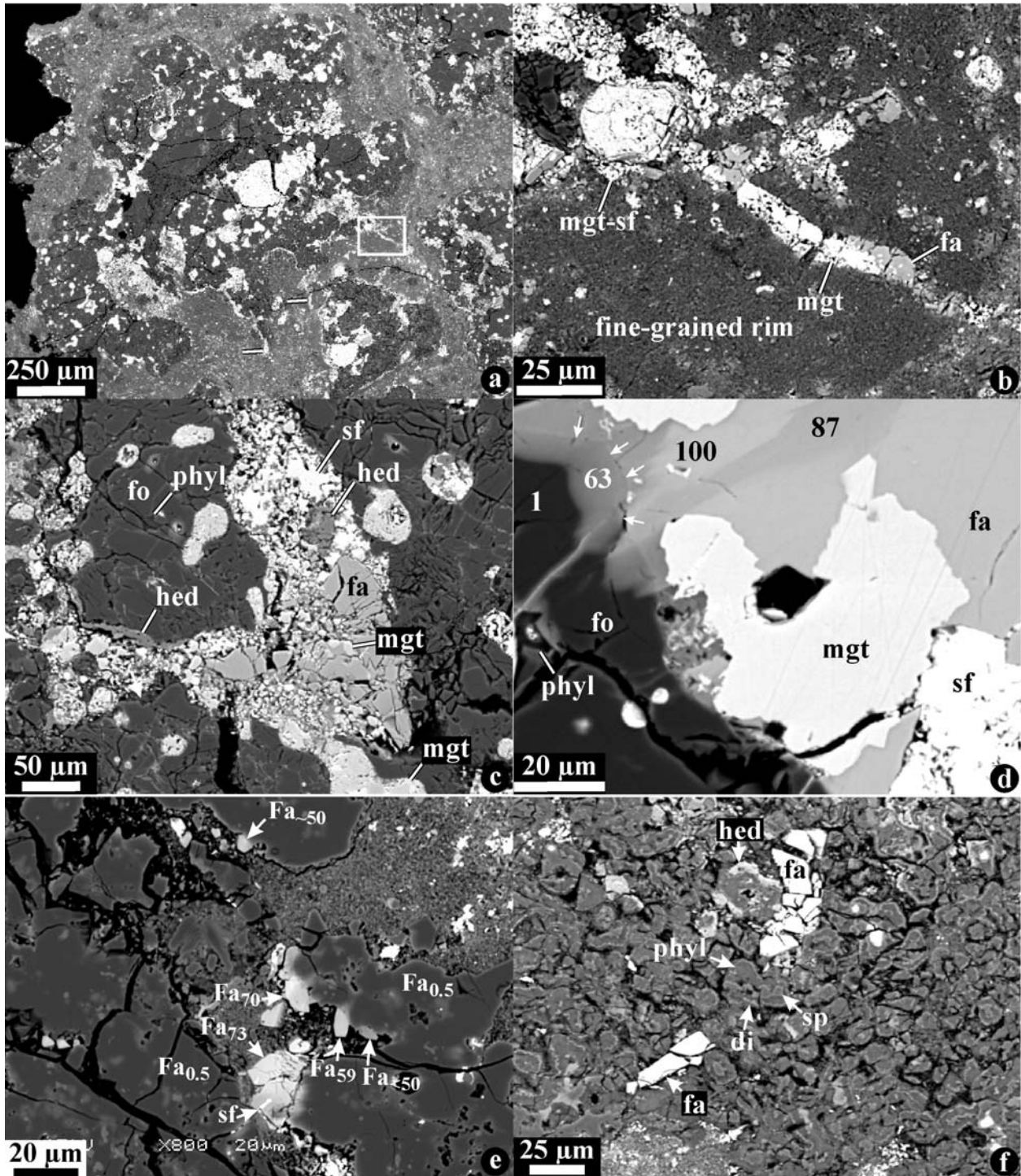


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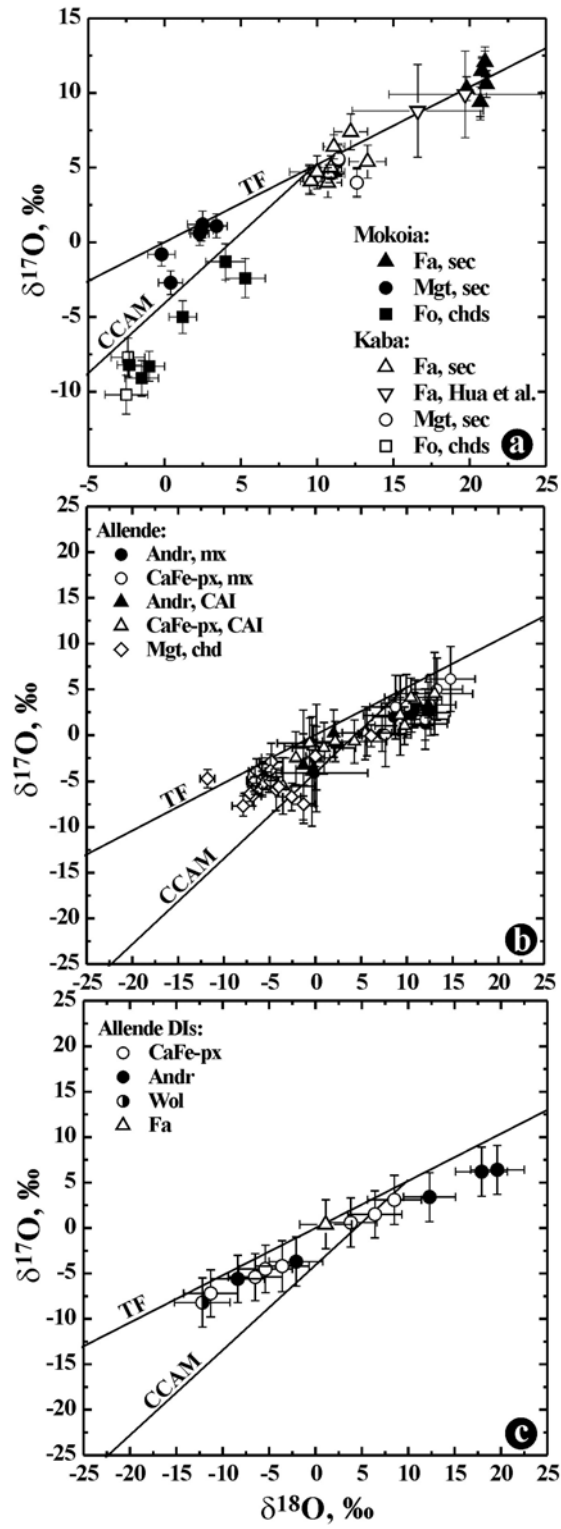


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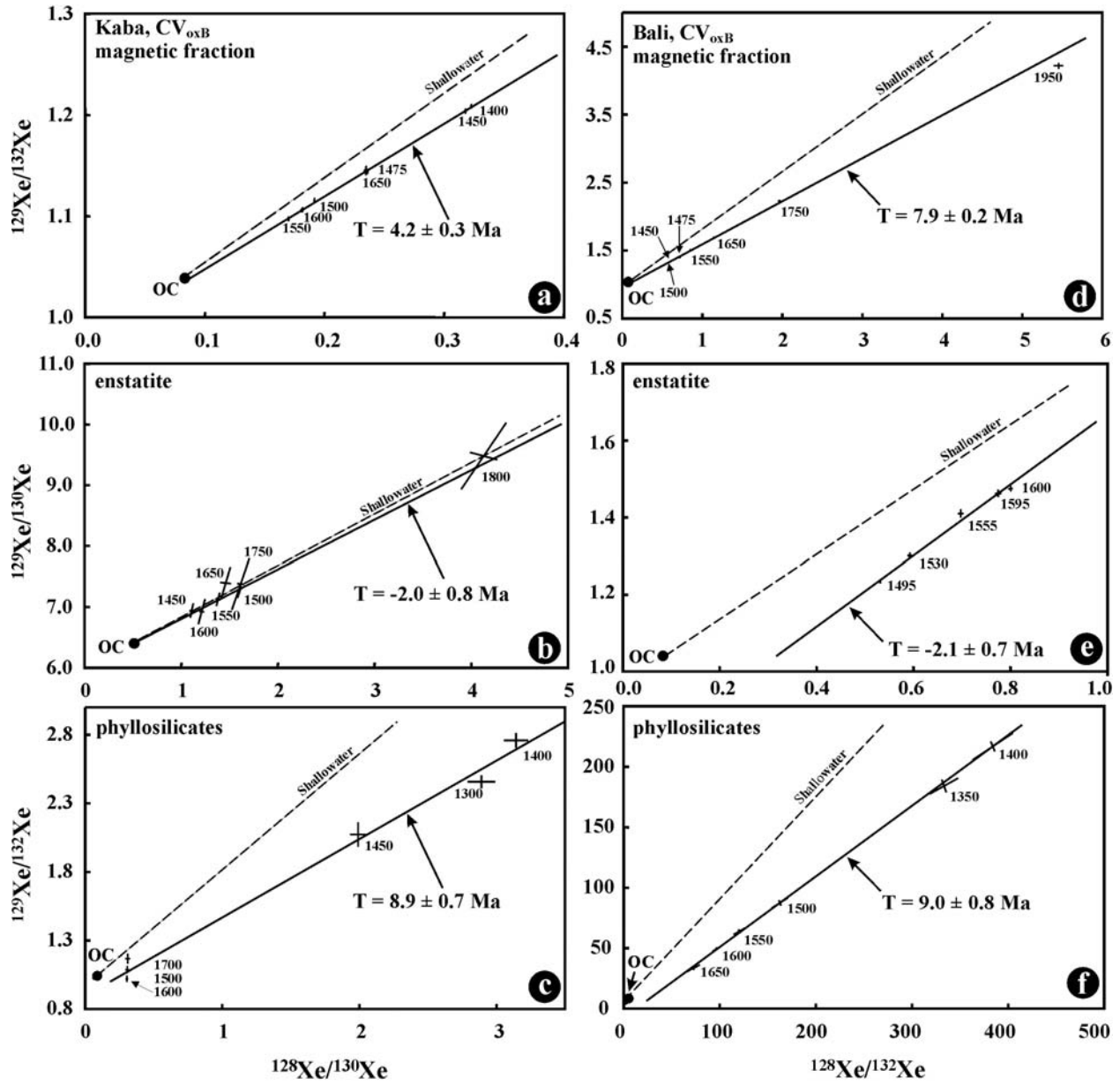


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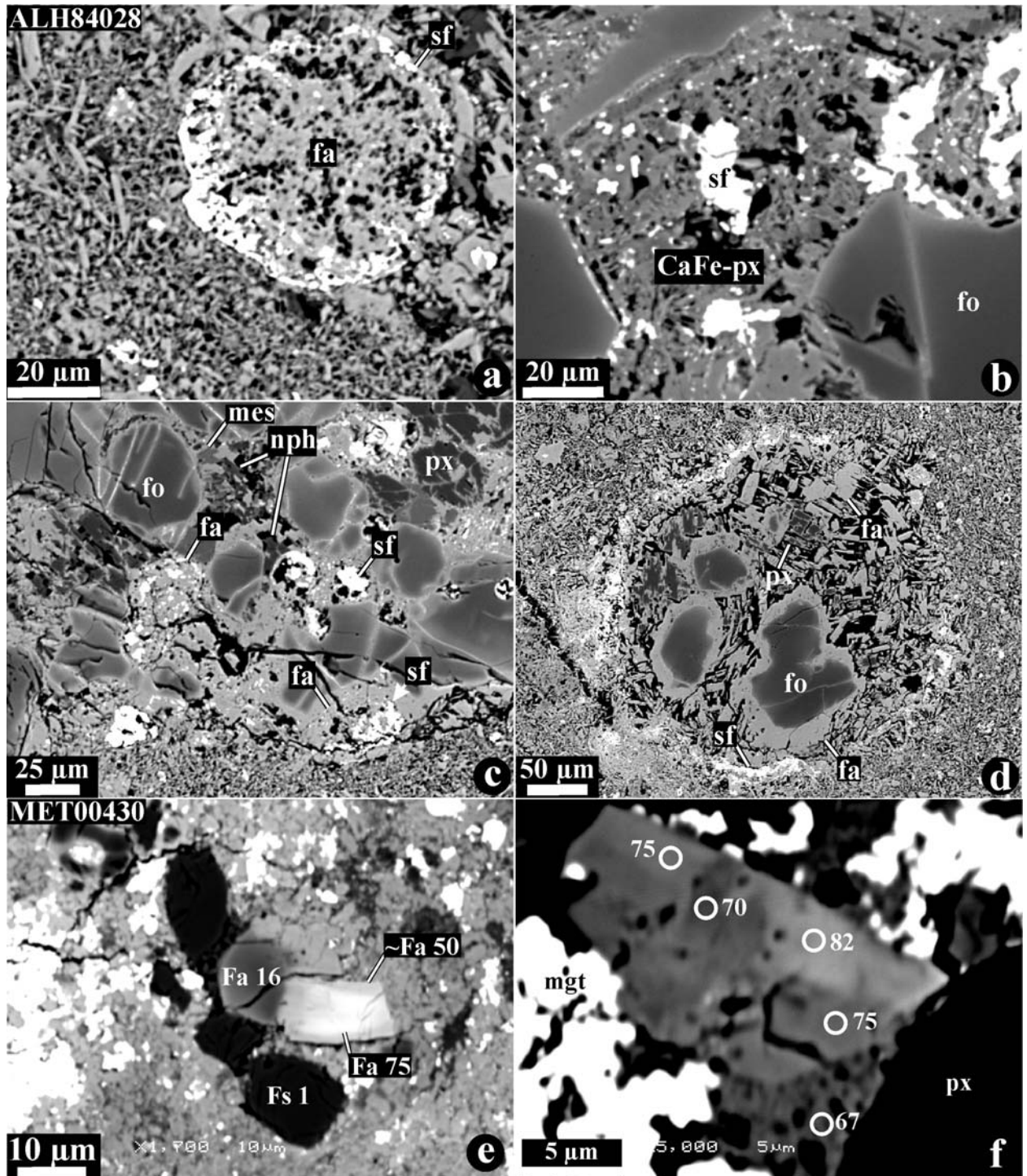


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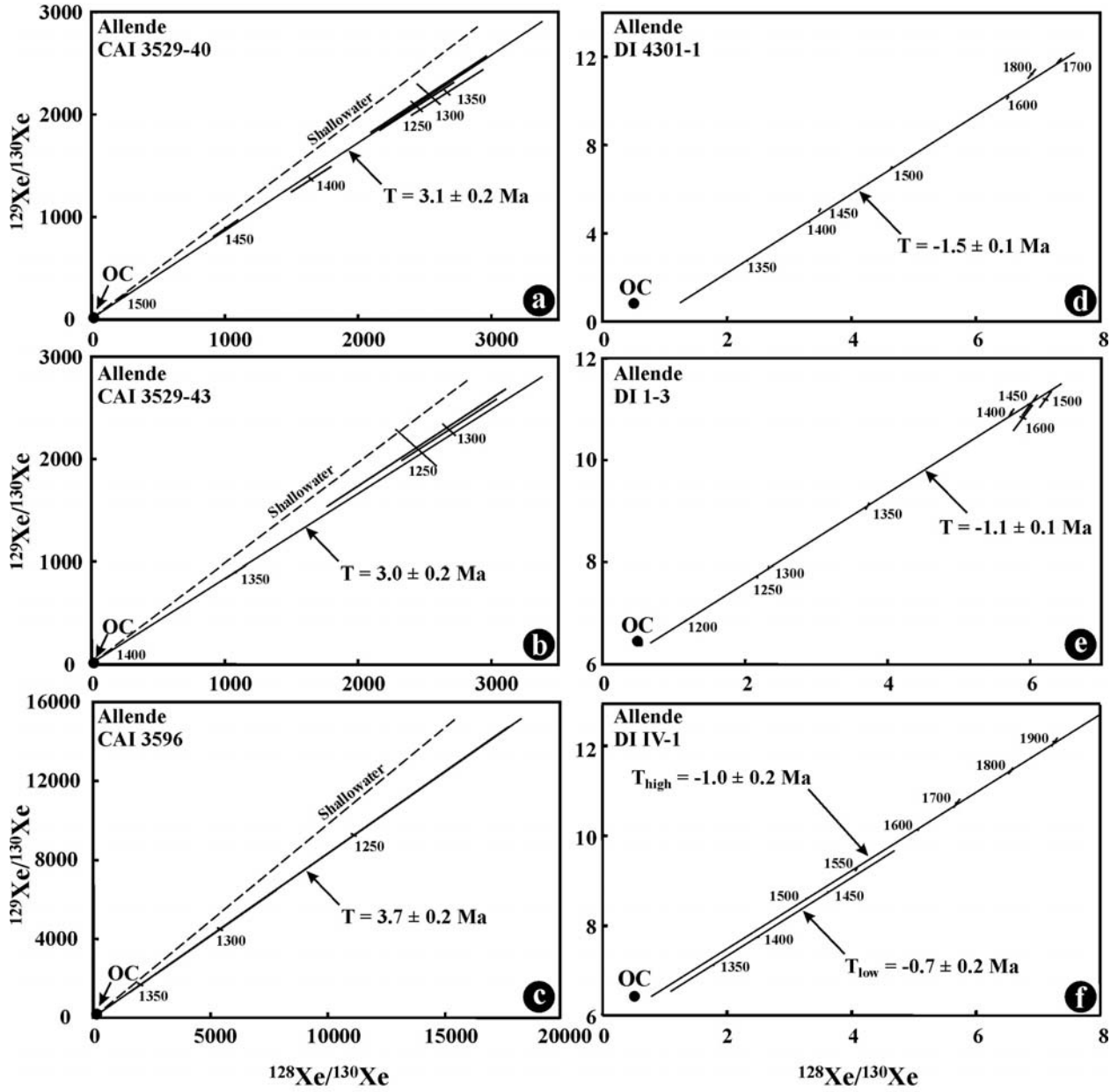


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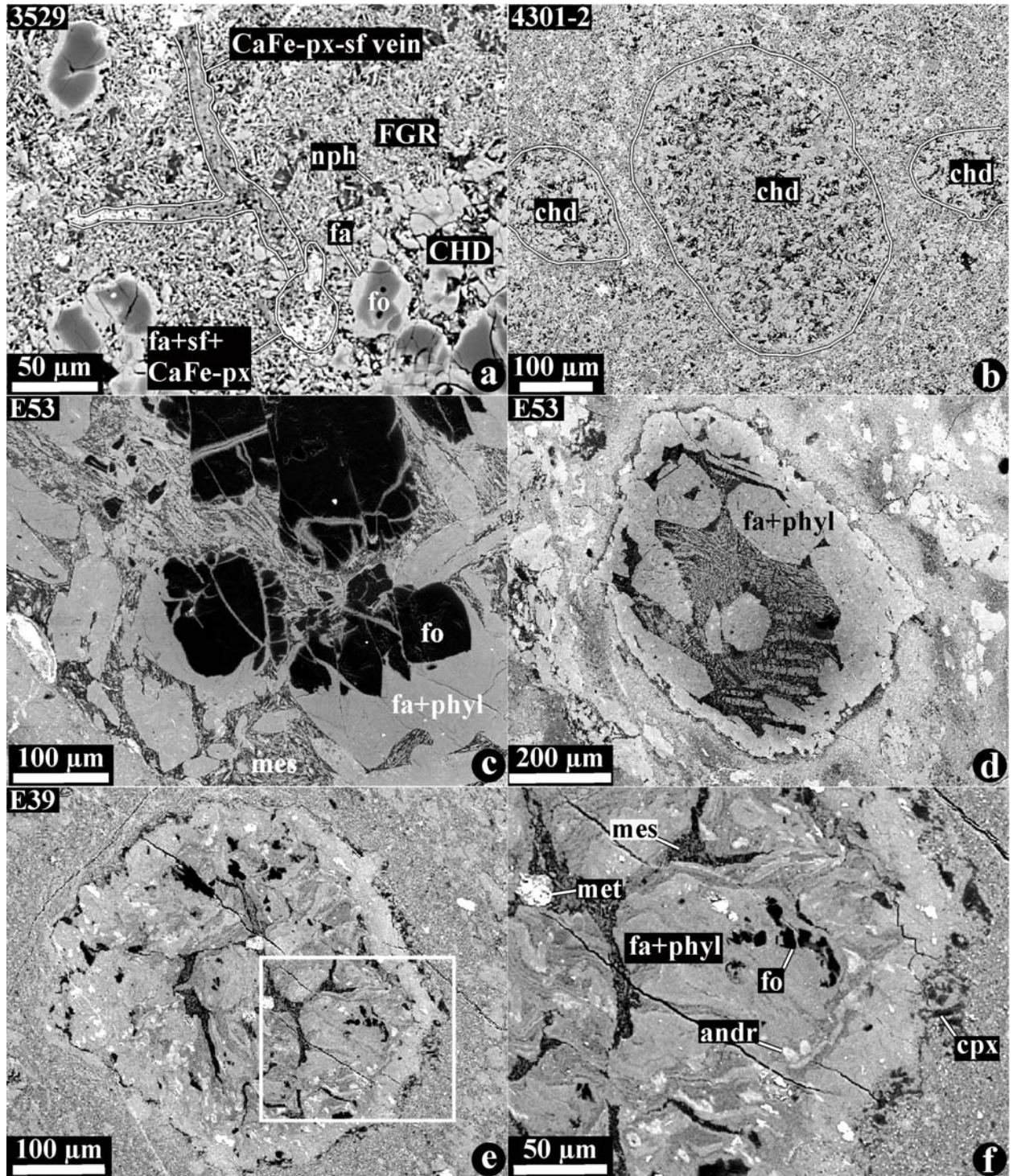


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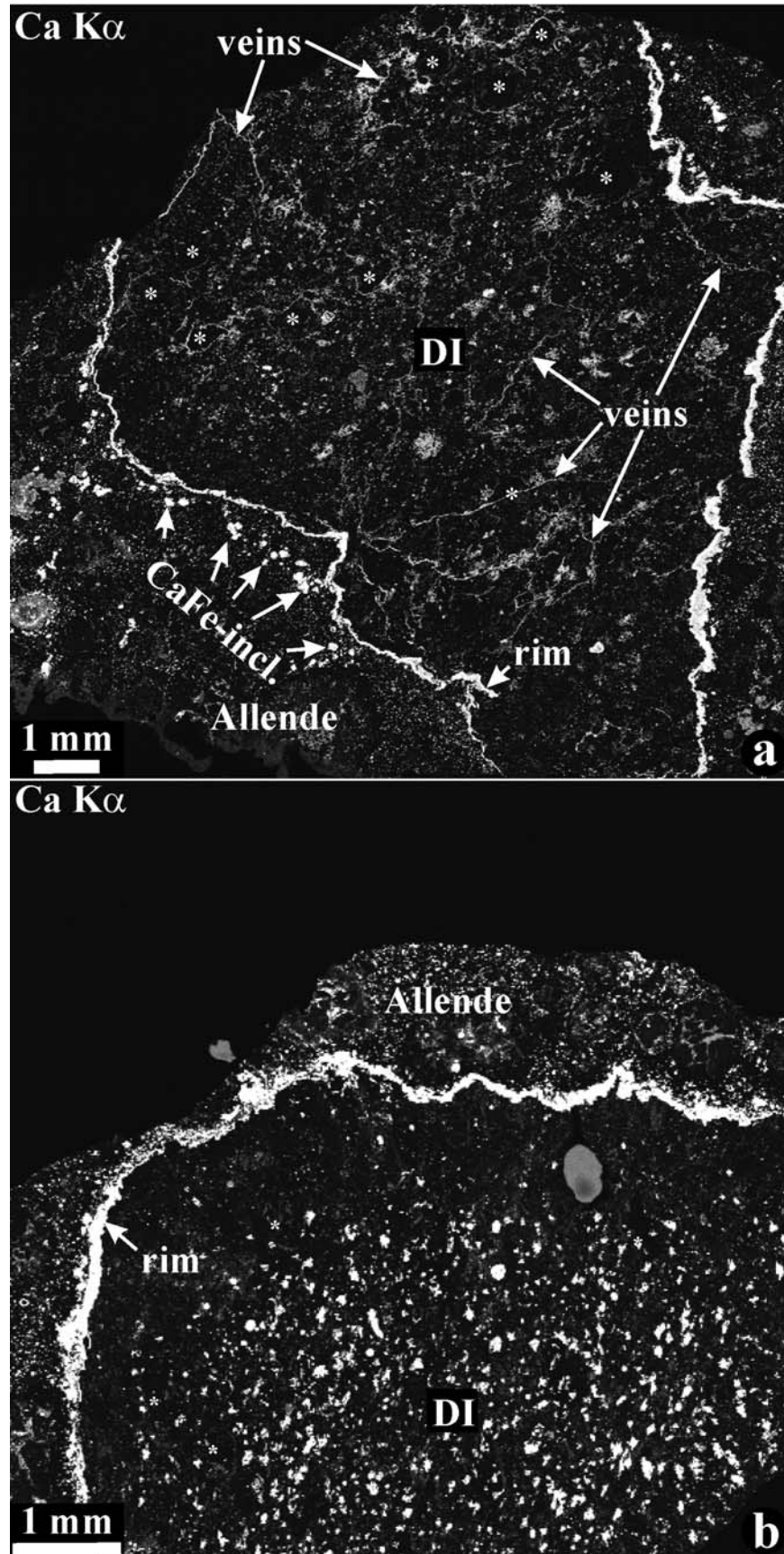


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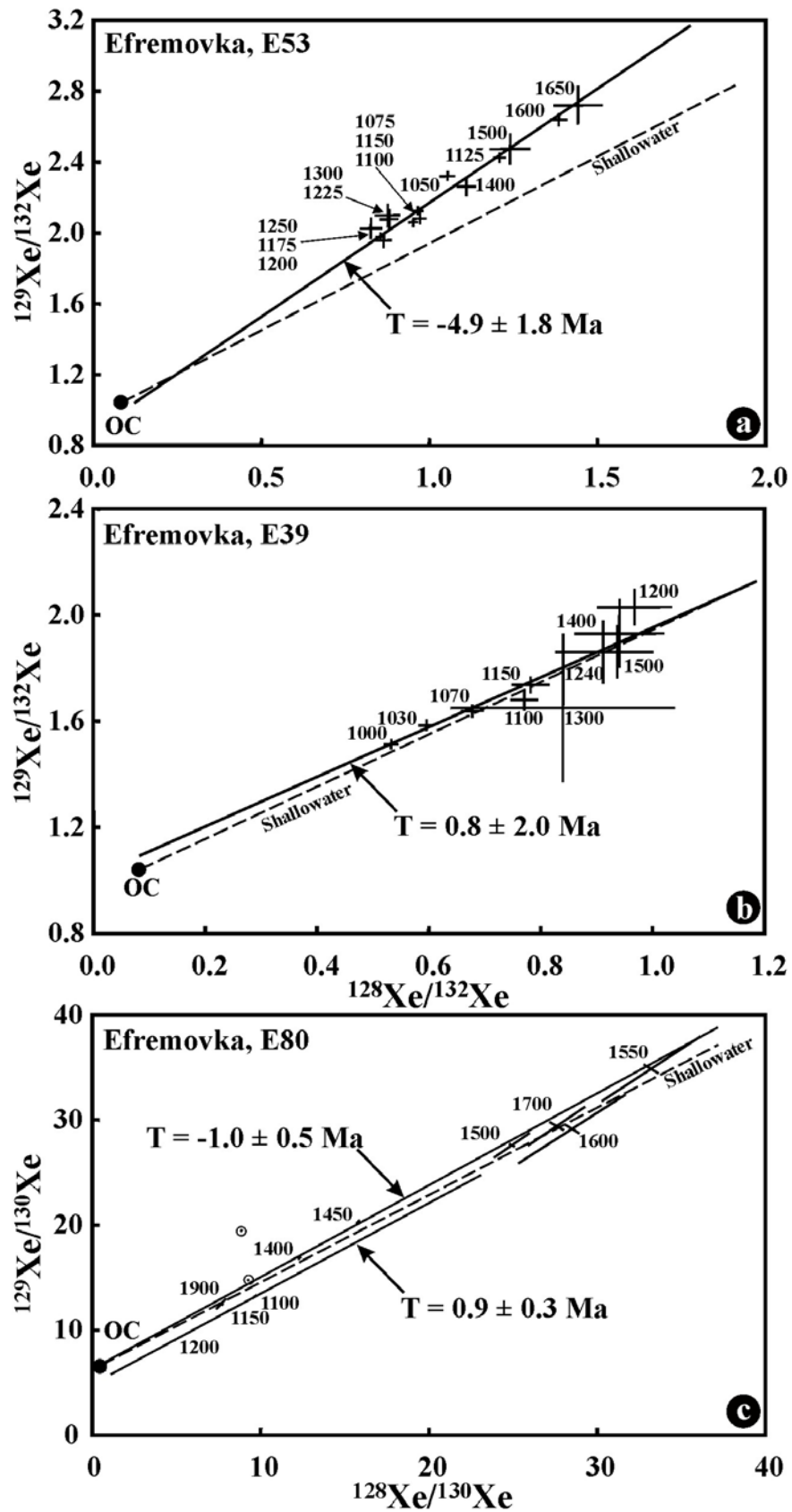


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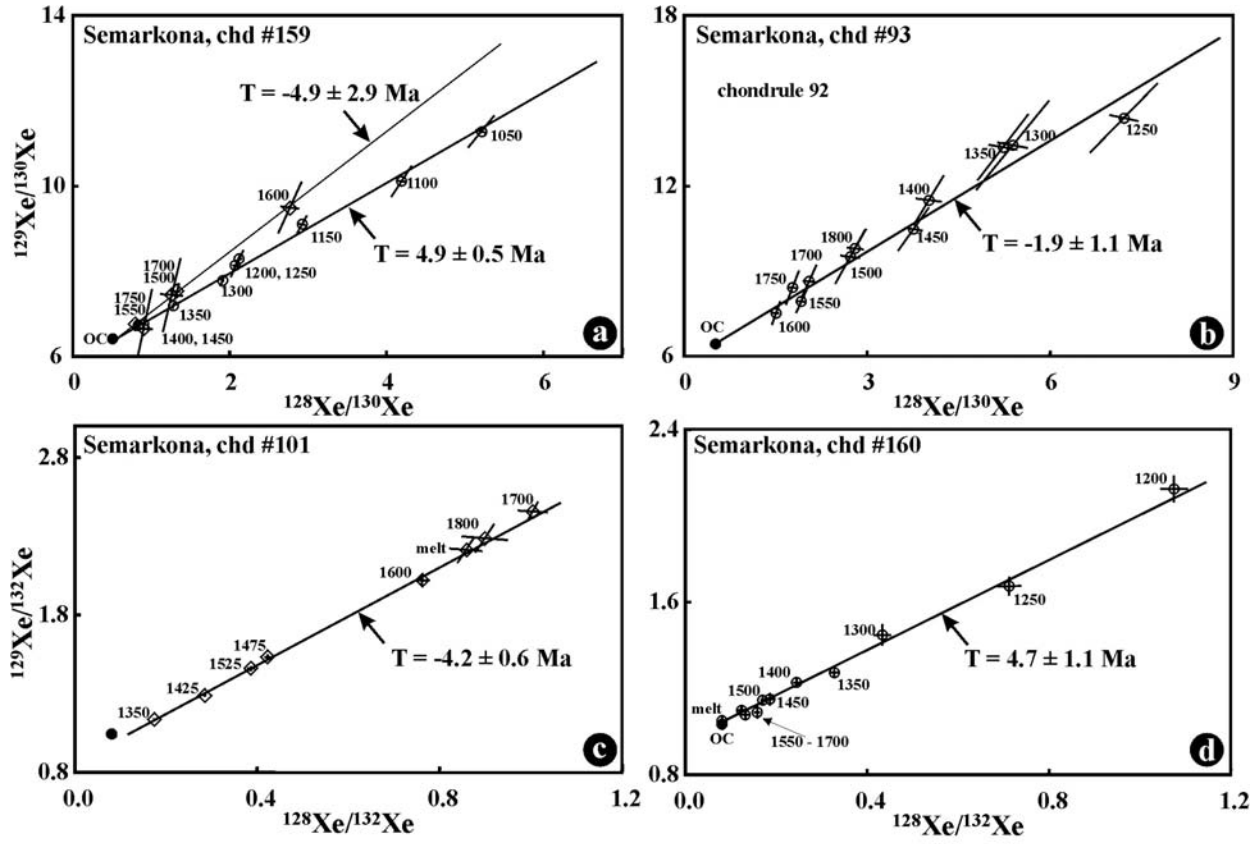


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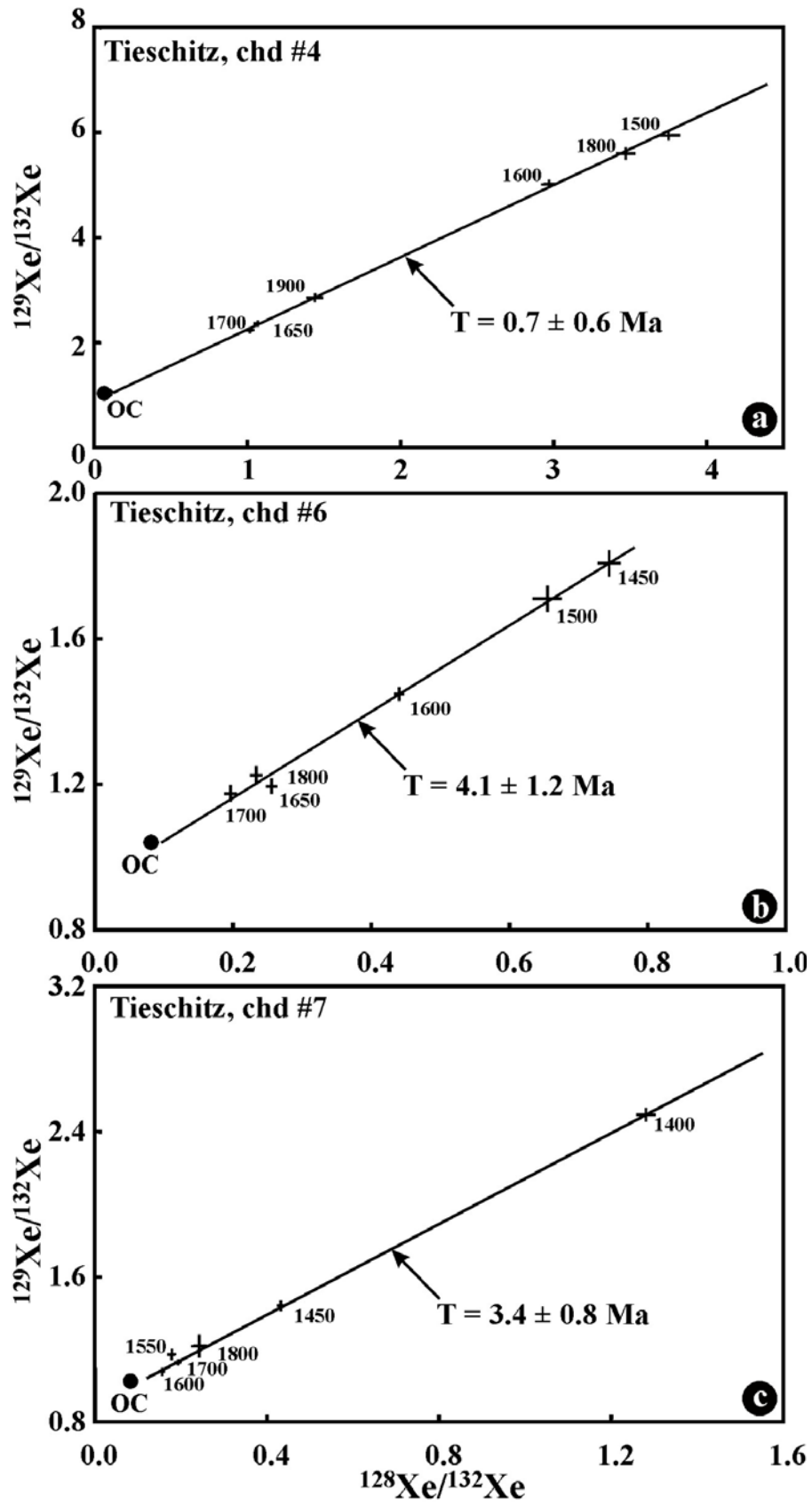


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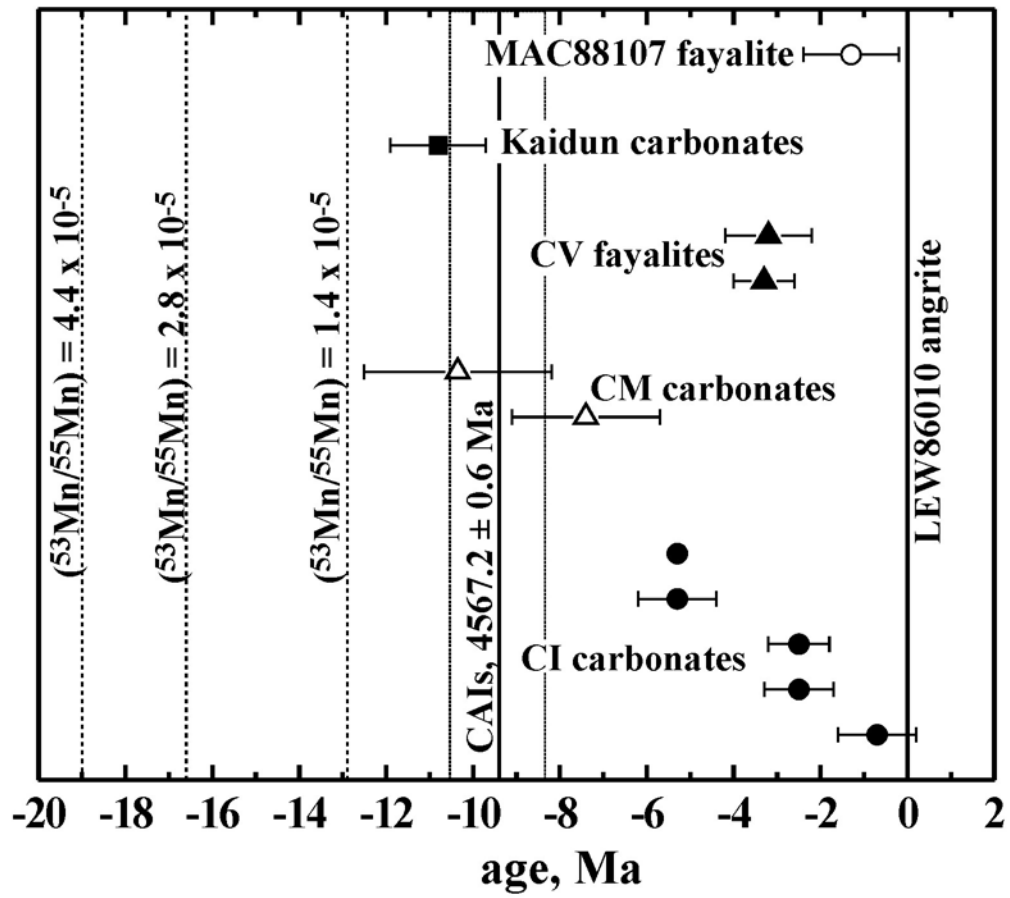


Fig. 19.

