



NASA Public Access

Author manuscript

Acta Geogr Geol Meteorol Debr Geol Gemorfol Termeszfoldr Sor. Author manuscript; available in PMC 2019 March 29.

Published in final edited form as:

Acta Geogr Geol Meteorol Debr Geol Gemorfol Termeszfoldr Sor. 2019 February 1; 246: 419–435. doi: 10.1016/j.gca.2018.11.002.

Evidence for oxygen isotopic exchange in chondrules from Kaba (CV3.1) carbonaceous chondrite during aqueous fluid-rock interaction on the CV parent asteroid

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Abstract

We report on the mineralogy, petrography, and oxygen isotopic compositions of primary olivine and plagioclase/feldspathic mesostases in chondrules and of secondary magnetite and fayalite in chondrules and matrix of an oxidized Bali-like CV3.1 carbonaceous chondrite, Kaba. In this meteorite, compositionally nearly pure fayalite (Fa_{98–100}) associates with hedenbergite (Fs_{~50}Wo_{~50}), magnetite, and Fe,Ni-sulfides. There are several textural occurrences of this mineral paragenesis: (i) coarse-grained intergrowths in interchondrule matrix, (ii) veins starting at the opaque nodules in the peripheries of type I chondrules and crosscutting finegrained rims around them, and (iii) rims overgrowing olivine of type I and type II chondrule fragments. Oxygen isotopic compositions of fayalite and magnetite are in disequilibrium with chondrule olivines. On a three-isotope oxygen diagram, $\delta^{17}\text{O}$ vs. $\delta^{18}\text{O}$, compositions of olivine plot along primitive chondrule minerals (PCM) line having a slope of ~ 1.0 ; deviations from the terrestrial fractionation line, $^{17}\text{O} = \delta^{17}\text{O} - 0.52 \times \delta^{18}\text{O}$, range from $\sim -8\%$ to $\sim -5\%$. In contrast, fayalite and magnetite plot along mass-dependent fractionation line with a slope of ~ 0.5 ; their $\delta^{18}\text{O}$ values range from -1 to $\sim +9\%$; ^{17}O is nearly constant (average $\pm 2\text{SE} = -1.5 \pm 1\%$). Oxygen isotopic compositions of chondrule plagioclase and feldspathic mesostases are in disequilibrium with chondrule olivines: they deviate to the right from the PCM line by $\sim 12\%$ and plot close to the mass-dependent fractionation line defined by fayalite and magnetite.

Based on the mineralogy, petrography, oxygen isotopic compositions of fayalite and magnetite, and the previously published thermodynamic analysis of the fayalite-bearing assemblages in ordinary and carbonaceous chondrites, we conclude that Kaba fayalite and magnetite formed during aqueous fluid-rock interaction at low water/rock ratio (0.1–0.2) and elevated temperatures (~ 200 – 300°C) on the CV chondrite parent asteroid. The ^{17}O values of Kaba fayalite and magnetite ($-1.5 \pm 1\%$) correspond to ^{17}O of aqueous fluid that operated on the CV chondrite parent asteroid and resulted in its alteration. Plagioclase and feldspathic mesostases in Kaba

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chondrules experienced postcrystallization oxygen isotopic exchange with this ^{16}O -depleted fluid; olivine grains retained their original compositions acquired during chondrule melts crystallization. The inferred oxygen isotopic exchange in Kaba chondrules appear to have not affected their Al-Mg isotope systematics.

Keywords

Kaba; chondrules; oxygen isotopes; fayalite; magnetite; oxygen-isotope exchange

Introduction and Previous Studies

Kaba is an oxidized Bali-like CV_{OXB}3.1 (Vigarano type) carbonaceous chondrite that experienced aqueous/hydrothermal alteration, but largely avoided subsequent thermal and/or shock metamorphism (e.g., KROT et al. 1998; BONAL et al. 2006). Nearly pure fayalite was first described in the CV_{OXB} chondrites, Kaba and Mokoia, by HUA & BUSECK (1995). Fayalite grains in these meteorites associate with magnetite, troilite, and pentlandite, reach up to 100 μm in size, and occur in the matrix, chondrules, and finegrained rims around chondrules and Ca,Al-rich inclusions (CAIs). HUA & BUSECK (1995) suggested fayalite in CV chondrites formed through nebular reaction of gaseous SiO, released by decomposition of enstatite, with magnetite and sulfides under highly oxidizing conditions, with $\text{H}_2\text{O}/\text{H}_2$ ratio much higher than the canonical solar nebula of $\sim 5 \times 10^{-4}$. Subsequently, based on the mineralogical observations, including the presence of fayalite-bearing veins crosscutting fine-grained rims around chondrules, thermodynamic analysis, and ^{53}Mn - ^{53}Cr dating, it was concluded that fayalite-bearing assemblages in Kaba and Mokoia formed during aqueous fluid-rock interaction $5.1_{-0.4}^{+0.5}$ Ma after formation of CV CAIs at ~ 200 – 300°C and low water-to-rock ratio (0.1–0.2) on the CV chondrite parent asteroid (e.g., KROT et al. 1998; CHOI et al. 2000; HUA et al. 2005; ZOLOTOV et al. 2006; BREARLEY & KROT 2013; DOYLE et al. 2015).

CHOI et al. (2000) and HUA et al. (2005) reported first relatively low-precision oxygen isotopic compositions (2σ for $\delta^{17}\text{O}$ and $\delta^{18}\text{O}$ ~ 3 – 4‰) of fayalite and magnetite in Kaba and Mokoia (Figs. 1a,b). Both groups found that on a three-isotope oxygen diagram, the compositions of magnetite and fayalite plot along a mass-dependent fractionation line close to the terrestrial fractionation (TF) line (average ^{17}O $\sim -0.2 \pm 1.5\text{‰}$, 2SE). There is, however, an important disagreement between these datasets: compositions of the Kaba fayalite and magnetite reported by HUA et al. (2005) have significantly different $\delta^{18}\text{O}$ than those reported by CHOI et al. (2000), -5 to $+10\text{‰}$ and $+17$ to $+24\text{‰}$ vs. $+10$ to $+13\text{‰}$ and $+10$ to $+13\text{‰}$, respectively. Assuming contemporaneous formation of magnetite and fayalite in equilibrium with an aqueous solution, these differences may significantly affect the temperature estimates of fayalite and magnetite formation (ZHENG 1991, 1993).

Recently, MARROCCHI et al. (2016) reported high-precision oxygen isotopic compositions (2σ for $\delta^{17}\text{O}$ and $\delta^{18}\text{O}$ ~ 0.5 – 1‰) of magnetite and olivine in Kaba chondrules (Figs. 1a,b). The magnetites show a range of $\delta^{18}\text{O}$ (-5 to 3.3‰) and ^{17}O (-3.7 to 0.1‰) and are in isotope disequilibrium with chondrule olivines (^{17}O $\sim -5\text{‰}$). On a three-isotope oxygen

diagram, the Kaba magnetites plot close to the primitive chondrule minerals (PCM) line and having a slope of ~ 1.0 (Fig. 1a). The PCM line was defined by high-precision oxygen isotopic measurements of chondrule silicates in Acfer 094 (USHIKUBO et al. 2012), which is one of the most primitive (C3.00) carbonaceous chondrites (GRESHAKE 1997). In contrast to previously reached conclusions (CHOI et al. 2000; HUE et al. 2005), MARROCCHI et al. (2016) suggested that magnetite in Kaba chondrules crystallized from chondrule melt under highly-oxidizing conditions. We note, however, that considering the observed range in $\delta^{18}\text{O}$ of $\sim 8\text{‰}$ and the typical uncertainties of ^{17}O of magnetite reported by MARROCCHI et al. (2016), $\sim 1.5\text{‰}$ (2σ), the limited range of ^{17}O does not allow us to distinguish the mass-independent and the mass-dependent fractionation lines (Figs. 1a,b). Oxygen isotopic compositions of fayalite, which appear to have formed contemporaneously with magnetite (e.g., KROT et al. 1998; DOYLE et al. 2005), could help to resolve this issue and provide additional constraints on the formation conditions of fayalite-magnetite-bearing assemblages.

In order to test the mechanism of magnetite formation proposed by MARROCCHI et al. (2016), to constrain oxygen-isotope composition of aqueous fluids on the CV parent asteroid, and to investigate a possible role of the fluid-rock interaction in oxygen isotopic exchange of high-temperature primary minerals in chondrules, we studied oxygen isotopic compositions of chondrule olivines and plagioclase/feldspathic mesostases, as well as fayalite and magnetite in chondrules and matrix from Kaba with the University of Hawai'i (UH) Cameca ims-1280 secondary ion mass-spectrometer (SIMS or ion microprobe).

Analytical procedures

The mineralogy and petrography of fayalite, magnetite, and chondrules in Kaba were studied in backscattered electron (BSE) images using the UHJXA-8500F field emission electron microprobe equipped with an energy dispersive spectrometer. Quantitative electron microprobe analyses were performed with JXA-8500F operated at 15 kV accelerating voltage, 15 nA beam current, and fully focused beam using five wavelength-dispersive spectroscopy detectors. For each element, counting times on both peak and background were 30 sec. Minerals with known chemical compositions were used as standards. A ZAF matrix correction method was used (a phi-rho-z correction; ARMSTRONG 1988). The element detection limits with these analytical conditions were (in wt%): Al_2O_3 and K_2O (0.01), SiO_2 , MgO , CaO , and Na_2O (0.02), TiO_2 , Cr_2O_3 , and MnO (0.03), and FeO (0.04).

Oxygen-isotope compositions of fayalite, magnetite, ferromagnesian olivines, and plagioclase/feldspathic mesostasis were measured *in situ* with the UH Cameca ims-1280 SIMS using a 15–20 pA focused Cs^+ primary ion beam of 20 keV impact energy and $\sim 2\ \mu\text{m}$ in diameter. The secondary ion mass spectrometer was operated at $-10\ \text{keV}$ with a 50 eV energy window. $^{16}\text{O}^-$ and $^{18}\text{O}^-$ were measured on multicollector Faraday Cup (FC) and electron multiplier (EM), respectively. $^{17}\text{O}^-$ was measured with the axial EM. The mass resolving power (m/m) for $^{16}\text{O}^-$ and $^{18}\text{O}^-$ was ~ 2000 , and that for $^{17}\text{O}^-$ was ~ 5500 , sufficient to separate interfering $^{16}\text{OH}^-$. A normal-incidence electron flood gun was used for charge compensation. To verify the positions of the sputtered regions, the spots analyzed for

oxygen isotopes were studied with BSE images using the UH JEOL JXA-8500F electron microprobe before and after SIMS measurements.

Oxygen-isotope compositions are reported as $\delta^{17}\text{O}$ and $\delta^{18}\text{O}$, deviations from Vienna Standard Mean Ocean Water (SMOW; $^{17}\text{O}/^{16}\text{O}_{\text{VSMOW}} = 0.000380$; $^{18}\text{O}/^{16}\text{O}_{\text{VSMOW}} = 0.002005$; DE LAETER et al. 2003) in parts per thousand: $\delta^{17,18}\text{O}_{\text{SMOW}} = [(^{17,18}\text{O}/^{16}\text{O}_{\text{sample}}) / (^{17,18}\text{O}/^{16}\text{O}_{\text{VSMOW}}) - 1] \times 1000$, and as ^{17}O .

Data for plagioclase/feldspathic mesostasis, ferramagnesian olivine, fayalite, and magnetite were corrected using Miyke-jima anorthite, San Carlos olivine, terrestrial fayalite and magnetite standards, respectively. The reported 2σ uncertainties include both the internal measurement precision on an individual analysis and the external reproducibility for standard measurements during a given analytical session. The external reproducibility on the multiple analyses of the standard was $\sim 1.5\text{--}2\%$ (2σ) for both $\delta^{17}\text{O}$ and $\delta^{18}\text{O}$.

Results

Mineralogy and petrography

Fayalite-magnetite±hedenbergite assemblages occur in all Kaba components — refractory inclusions, chondrules, and matrix, and have been previously characterized in detail (HUA & BUSECK 1995; KROT et al. 1998; this study). In *Figures 2* and *3a,b*, we illustrated typical occurrences of Kaba fayalite and magnetite, including (i) coarse fayalite grains inside type I chondrules (*Figs. 2a,b*), (ii) fayalite-magnetite-sulfide veins crosscutting fine-grained rims around chondrules (*Figs. 2c,d*), (iii) fayalite±hedenbergite overgrowths on chondrule fragments (*Fig. 2e*), and (iv) coarse fayalite grains in the matrix (*Fig. 2f*). All textural occurrences of fayalite grains measured are nearly pure Fe_2SiO_4 ($\text{Fa}_{>98}$)

There are two major textural occurrences of magnetite in Kaba: magnetite-sulfide nodules replacing Fe,Ni-metal in type I chondrules, and massive subhedral magnetite grains, often with elongated inclusions of Fe,Ni-sulfides, overgrowing magnetite-sulfide nodules (*Figs. 3a,b*). Nodular magnetite contains high chromium contents (up to 1 wt%), whereas massive magnetite is compositionally pure FeFe_2O_4 .

Plagioclase and glassy mesostasis in magnetite±fayalite-bearing chondrules are replaced to various degrees by phyllosilicates and unidentified FeO-rich secondary minerals (*Figs. 3b–f*).

Oxygen isotopic compositions

Oxygen isotopic compositions of chondrule olivines, chondrule plagioclase/feldspathic mesostasis, fayalite, and magnetite are plotted in *Figures 1c* and *d*. Chondrule olivines show a range of ^{17}O (-7.6 to -4.4%) and on a three-isotope oxygen diagram plot along the PCM line. The similar results for chondrule olivine and low-Ca pyroxene phenocrysts in Kaba were reported by HERTWIG et al. (2016). In contrast to isotopically uniform chondrules from Acfer 094 (USHIKUBO et al. 2012), the plagioclase/feldspathic mesostasis in Kaba chondrules

are in oxygen-isotope disequilibrium with olivine phenocrysts; the former plot significantly to the right from the PCM line and have ^{17}O of $-0.5\pm 0.9\text{‰}$.

All three major textural occurrences of fayalite analyzed (matrix, chondrule overgrowths and coarse grains inside chondrules) have similar oxygen isotopic compositions: $\delta^{18}\text{O}$ range from ~ -7 to $\sim 9\text{‰}$; the average ^{17}O is $\sim -1.5\pm 1.3\text{‰}$ (2SE). Massive magnetite grains in type I chondrules have average ^{17}O $\sim -1.2\pm 1.1\text{‰}$, and a relatively small range of $\delta^{18}\text{O}$, from ~ -1 to $\sim +6\text{‰}$. On a three-isotope oxygen diagram (Fig. 1c), the compositions of fayalite and magnetite plot along a single mass-dependent fractionation line with ^{17}O of $\sim -1.5\text{‰}$. Within uncertainty of our measurements, compositions of chondrule plagioclase/mesostasis plot along this line as well (Figs. 1c,d).

Discussion

Oxygen isotopic exchange during aqueous fluid-rock interaction

Mineralogical observations and thermodynamic analysis suggest that fayalite and magnetite in Kaba resulted from fluid-assisted metasomatic alteration at $\sim 200\text{--}300^\circ\text{C}$ on the CV parent asteroid (KROT et al. 1998; ZOLOTOV et al. 2006; BREARLEY & KROT 2013; DOYLE et al. 2015). Since these minerals formed by oxidation of Fe,Ni-metal and/or by direct precipitation from an aqueous fluid (KROT et al. 1998; BREARLEY & KROT 2013), their ^{17}O of $-1.5\pm 1\text{‰}$ must correspond to ^{17}O of the fluid (KROT et al. 2015).

In most chondrules from the least metamorphosed chondrites [petrologic type < 3.0 , e.g., Acfer 094 (C3.0 ungrouped), Yamato-81020 (CO3.0), and CR2-3s], olivine and low-Ca pyroxene phenocrysts, plagioclase, and glassy mesostases are in oxygen isotopic equilibrium, i.e., within an individual chondrule, these minerals have the same ^{17}O (within uncertainty of SIMS measurements), suggesting crystallization from a melt having nearly constant ^{17}O (e.g., USHIKUBO et al. 2012; TENNER et al. 2013, 2015). In contrast, olivine phenocrysts and plagioclase/feldspathic mesostasis in Kaba chondrules are in oxygen isotopic disequilibrium (Figs. 1c,d), indicating postcrystallization isotope exchange that affected only the plagioclase/feldspathic mesostasis. On a three-isotope oxygen diagram (Fig. 1c), the olivine phenocrysts plot along the PCM line, whereas the plagioclase grains deviate to the right from the PCM line and plot along mass-dependent fractionation line with ^{17}O of $\sim -1.5\text{‰}$ defined by the Kaba magnetite and fayalite, suggesting the exchange resulted from interaction with the fluid. Plagioclase/feldspathic mesostasis in Kaba chondrules have nearly the same (within uncertainty of our SIMS measurements) ^{17}O as magnetite and fayalite, indicating that the exchange was nearly complete.

It has been previously shown that the presence of water leads to significantly enhanced oxygen-diffusion rates in many silicates compared to those under dry conditions (e.g., FARVER 2010 and references therein). In plagioclase, oxygen-diffusion coefficient under wet (hydrothermal) conditions (GILETTI et al. 1978) is considerably higher than that under dry conditions (YURIMOTO et al. 1989; RYERSON & MCKEEGAN 1994) (Fig. 4a). Under the inferred temperature of metasomatic alteration experienced by Kaba, $200\text{--}300^\circ\text{C}$ (ZOLOTOV et al. 2006), a $10\ \mu\text{m}$ -sized crystal of anorthite (typical grain sizes of plagioclase in Kaba

chondrules) would completely exchange its oxygen isotopic composition with an aqueous fluid within ~0.1–0.001 Ma (Fig. 4b). This time scale may not be unreasonable for a duration of alteration on the CV parent asteroid. We note, however, that the duration for the presence of aqueous fluid on the CV parent asteroid is still poorly constrained.

Conclusions

We reported on the mineralogy, petrography, and *in situ* measured oxygen-isotope compositions of primary high-temperature minerals in chondrules (olivines and plagioclase/feldspathic mesostasis) and of secondary fayalite and magnetite in chondrules and matrix of the Kaba (CV_{oxB}3.1) carbonaceous chondrite.

Oxygen isotopic compositions of fayalite and magnetite appear to have equilibrated with external reservoir(s) having nearly the same ¹⁷O: on a three-isotope oxygen diagram, their compositions plot along mass-dependent fractionation line with ¹⁷O of ~–1.5‰. Magnetite and fayalite are in isotope disequilibrium with chondrule olivines, which plot along a slope-1 line and show a range of ¹⁷O, from ~–8‰ to ~–5‰. These data and the observed spread of δ¹⁸O of magnetite and fayalite (from –1 to +9‰) are inconsistent with high-temperature formation of magnetite during crystallization of chondrule melts (MARROCCHI et al. 2016), instead, they are consistent with formation of magnetite and fayalite during relatively low-temperature aqueous fluid-rock interaction (KROT et al. 1998; CHOI et al. 2000; DOYLE et al. 2017). Compositions of chondrule plagioclase/feldspathic mesostases are in isotopic disequilibrium with chondrule olivines: their ¹⁷O are similar to those of fayalite and magnetite.

Based on the mineralogy, petrography, oxygen isotopic compositions of fayalite and magnetite, and thermodynamic analysis of the fayalite-bearing assemblages in chondrites (ZOLOTOV et al. 2006), we conclude that fayalite and magnetite in Kaba formed during aqueous fluid-rock interaction at low water/rock ratio (0.1–0.2) and elevated temperatures (~200–300°C) on the CV chondrite parent asteroid. Therefore, the ¹⁷O values of the fayalite and magnetite correspond to ¹⁷O of aqueous fluid that operated on the CV chondrite parent asteroid and resulted in its alteration. Plagioclase and feldspathic mesostasis in Kaba chondrules experienced postcrystallization oxygen isotopic exchange with this aqueous fluid; olivine grains retained their original compositions. The inferred oxygen isotopic exchange in Kaba chondrules appear to have not affected their Al-Mg isotope systematics (NAGASHIMA et al. 2017).

Acknowledgements:

We would like to thank all organizers of the Conference at the University of Debrecen. We thank Dr. S. S. Russell (Natural History Museum, London), Dr. M. Gounelle (Natural History Museum, Paris), Dr. M. I. Petaev (Smithsonian Institution), and Dr. M. Wadhwa (Arizona State University) for providing polished thin sections of Kaba. We thank Drs. A. E. Rubin and J. T. Wasson for their comments and suggestions which helped to improve the paper. Handling the paper by Dr. R. McIntosh is highly appreciated. This work was supported by the NASA Emerging Worlds program grant NNX17A22G (A. N. Krot, P.I.).

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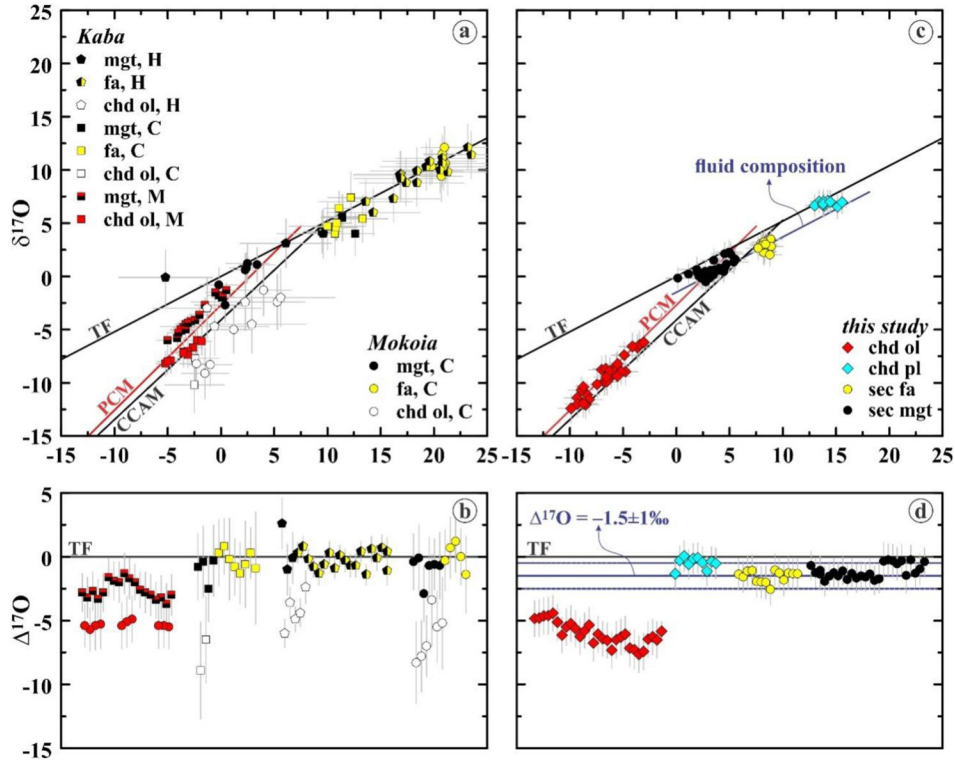


Figure 1:

(a, c) Three-isotope oxygen diagram and (b, d) ^{17}O values of the chondrule olivine phenocrysts (chd ol), chondrule plagioclase/feldspathic mesostasis (chd pl), and secondary fayalite (secfa) and magnetite (sec mgt) from the Bali-like oxidized CV3 chondrites, Kaba and Mokoia. (a, b) — Previously published data from (CHOI et al 2000) — (C), (HUA et al 2005) — (H), and (MARROCCHI et al. 2016) — (M). (c, d) — this study. Carbonaceous chondrite anhydrous mineral (CCAM) line (CLAYTON et al. 1977), primitive chondrule minerals (PCM) line (USHIKUBO et al. 2012), and the terrestrial fractionation (TF) line are shown for reference. Chondrule olivines plot along the PCM line; magnetite and fayalite plot along mass-dependent fractionation line with ^{17}O of $\sim -1.5\text{‰}$, which corresponds to ^{17}O of an aqueous fluid resulting on formation of fayalite and magnetite. Compositions of chondrule olivines are in isotope disequilibrium with chondrule plagioclase/feldspathic mesostases; the latter, however, are in isotopic equilibrium with secondary fayalite and magnetite, suggesting oxygen isotopic exchange with the aqueous fluid

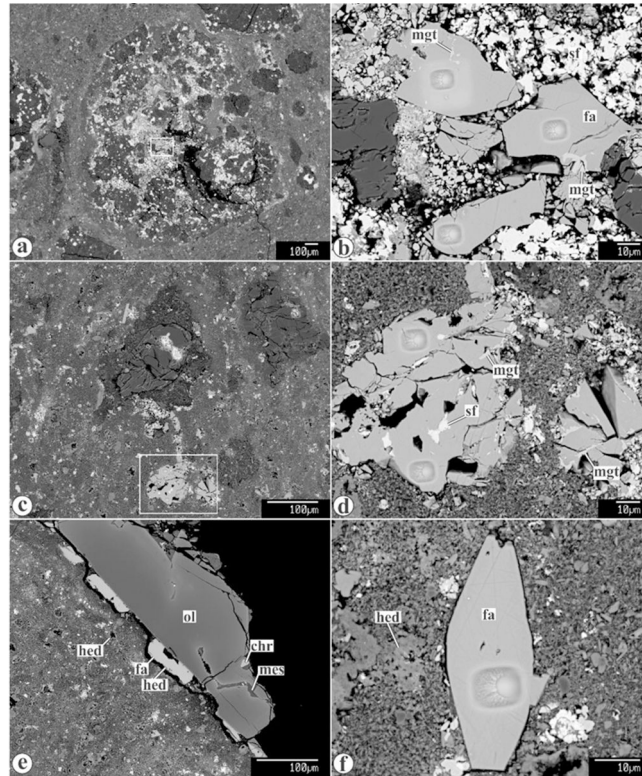


Figure 2: Backscattered electron images of textural occurrences of secondary fayalite in Kaba. chr = chromite; fa = fayalite; hed = hedenbergite; mes = mesostasis; mgt = magnetite; ol = ferromagnesian olivine; sf = sulfide. Fayalite associated with magnetite, hedenbergite, and sulfides (a, b) replaces Fe,Ni-metal nodules in type I chondrules, (c, d) forms veins crosscutting fine-grained rims around chondrules, (e) overgrows chondrule olivines, and (f) occurs as euhedral grains in the matrix.

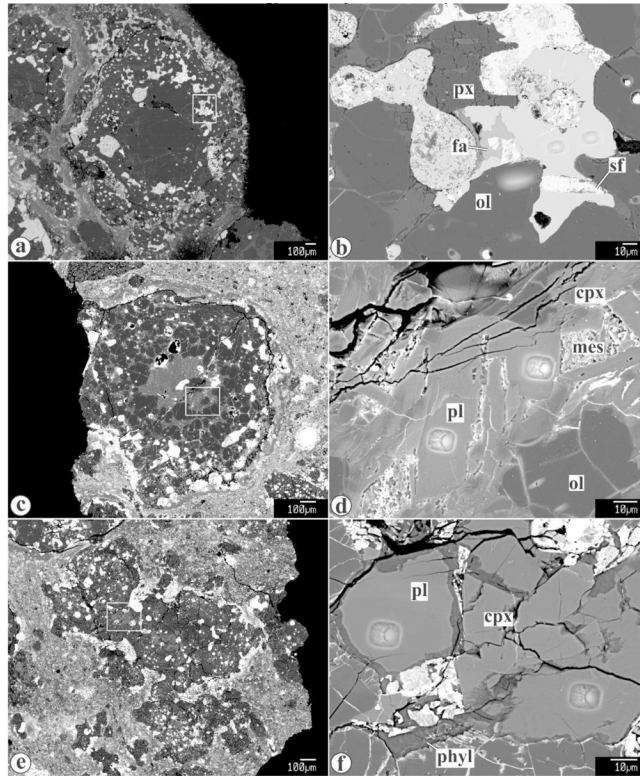


Figure 3: Backscattered electron images of Kaba chondrules with magnetite, fayalite, plagioclase and olivine phenocrysts measured for oxygen-isotope compositions. Regions outlined in (a, c, and d) are shown in detail in (b, d, and f, respectively). cpx = high-Ca pyroxene; fa = fayalite; mes = mesostasis; mgt = magnetite; ol = olivine; pl = plagioclase/feldspathic mesostasis; phyl = phyllosilicates; px = low-Ca pyroxene; sf = Fe,Ni-sulfides.

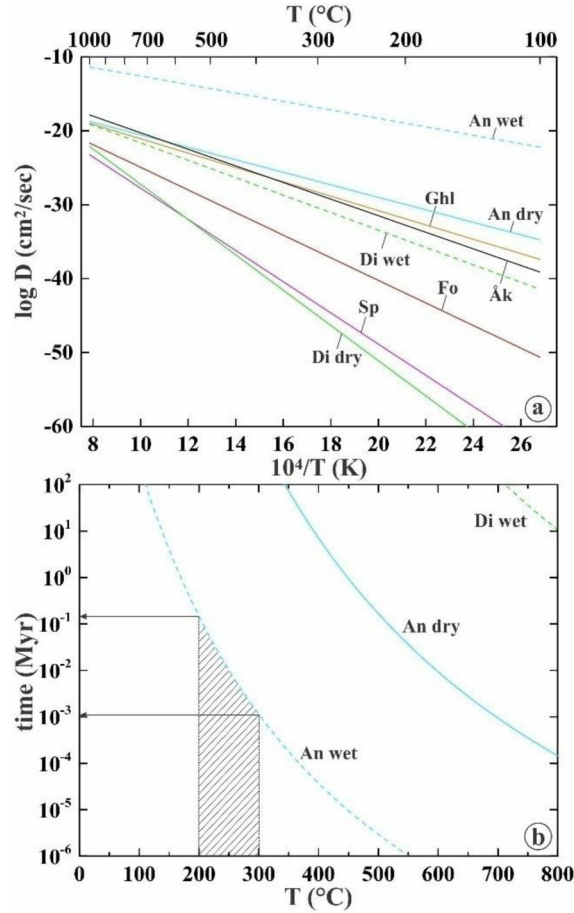


Figure 4:
 (a) Arrhenius plot of oxygen diffusion coefficients in spinel, åkermanite, gehlenite, diopside, forsterite, and anorthite under dry and wet conditions. Data from GILETTI et al (1978), OISHI & ANDO (1984), FARVER (1989), YURIMOTO et al (1989), and RYERSON & MCKEEGAN (1994). (b) Temperature and time dependence for a complete oxygen isotopic exchange in a 10 µm-sized anorthite (typical size of plagioclase grains in Kaba chondrules) under dry (red line) and wet (blue line) conditions. Under the estimated conditions of metasomatic alteration of Kaba (200–300°C), it takes between 0.1 and 0.001 Ma to exchange oxygen isotopic composition of plagioclase with an aqueous fluid.