

^{53}Mn - ^{53}Cr Dating of Fayalite Formation in the CV3 Chondrite Mokoia: Evidence for Asteroidal Alteration

Ian D. Hutcheon,* Alexander N. Krot, Klaus Keil,
Douglas L. Phinney, Edward R. D. Scott

Fayalite grains in chondrules in the oxidized, aqueously altered CV3 chondrite Mokoia have large excesses of radiogenic chromium-53. These excesses indicate the in situ decay of short-lived manganese-53 (half-life = 3.7 million years) and define an initial $^{53}\text{Mn}/^{55}\text{Mn}$ ratio of $2.32 (\pm 0.18) \times 10^{-6}$. This ratio is comparable to values for carbonates in CI and CM chondrites and for several classes of differentiated meteorites. Mokoia fayalites formed 7 to 16 million years after Allende calcium-aluminum-rich inclusions, during hydrothermal activity on a geologically active asteroid after chondritic components had ceased forming in the solar nebula.

Chondrites, which consist of chondrules, Ca-Al-rich inclusions (CAIs), and matrix material, are a class of meteorites thought to represent remnants of primordial solid particles formed in the solar nebula (1). The chemical, isotopic, and petrologic properties of the minerals in chondrites have been used to infer the chemical and physical processes operating in the solar nebula. Unfortunately, chondrites are not pristine relics but were heated and altered during metamorphism, after their primary constituents had accreted into asteroids (2). Metamorphism may overprint the chemical and textural record of early solar system processes; initially, studies focused on carbonaceous chondrites (3), such as Allende and other members of the CV3 group, because these meteorites appeared relatively free of secondary minerals produced during metamorphism and aqueous alteration (4). However, recent work suggests that components in Allende and most other CV3 chondrites were altered after they formed (5–8). The question remains open as to where and when these secondary minerals formed: in the solar nebula before the chondrules, CAIs, and matrix material accreted, or on planetesimals (or CV3 asteroids) after accretion (9).

The extent and nature of the secondary mineralogical changes in CV3 chondrites vary widely among the oxidized and reduced subgroups of these chondrites. The reduced subgroup (10) exhibits minimal alteration; type I (MgO-rich) chondrules, the most abundant

type, contain largely unaltered, rounded nodules of metallic FeNi and troilite (FeS) and glassy mesostases. In the Bali-like oxidized subgroup (11), chondrules contain secondary phyllosilicates, magnetite, Ni-rich metal, pentlandite [(Fe, Ni) $_9$ S $_8$], fayalite (Fa $_{90-100}$), and Ca-Fe-rich pyroxenes varying in composition from Fs $_{10}$ Wo $_{50}$ to Fs $_{50}$ Wo $_{50}$ (where Fs is ferrosilite and Wo is wollastonite) (12). In the Allende-like oxidized subgroup (11), fayalite and phyllosilicates are rare or absent (13, 14) and secondary fayalitic olivine (Fa $_{30-50}$), nepheline, and sodalite are present instead. Secondary minerals in the oxidized subgroups formed either in the solar nebula by gas-solid reactions above 800°C in an oxidizing environment (8, 15) or in an asteroid at low temperatures (<300°C) during aqueous or hydrothermal alteration (5, 6).

Because metamorphism and alteration in asteroids continued long after the solar nebula had dissipated, determining the time of formation of secondary minerals in chondrites can potentially distinguish between nebular and asteroidal alteration. Three short-lived, and now extinct, radionuclides— ^{26}Al , ^{53}Mn , and ^{129}I , with respective half-lives of 0.75, 3.7, and 15.7 million years (My)—have been used to date secondary minerals (16, 17). The absence of radiogenic ^{26}Mg , produced by the decay of ^{26}Al , in feldspathoids and grossular in most Allende CAIs suggests that alteration occurred at least 2 My after CAI formation (16), whereas I-Xe ages of Allende feldspathoids indicate that these minerals formed at least 4 My after CAI crystallization (17). Carbonates in CI (Ivuna, Orgueil) and CM (Nogoya) carbonaceous chondrites, which formed by aqueous alteration on their parent asteroids (18), contain excesses of radiogenic ^{53}Cr with inferred initial $^{53}\text{Mn}/^{55}\text{Mn}$ values of $\sim 2.0 \times 10^{-6}$ (19, 20). Similar

initial $^{53}\text{Mn}/^{55}\text{Mn}$ ratios are inferred for five groups of differentiated meteorites: angrites, pallasites, IIIAB irons, noncumulate eucrites, and brachinites (21). These data suggest that aqueous alteration on the CI and CM asteroids occurred nearly simultaneously with igneous differentiation on other asteroids, ~ 5 to 20 My after CAI formation. The isotopic evidence for an extended time scale of secondary mineralization favors an asteroidal setting, but a nebular origin of some metamorphic features cannot be excluded (22).

The fayalite grains occurring in the Bali-like oxidized subgroup of CV3 chondrites are large (up to 100 μm in size) and have high Mn/Cr ratios ($>10^3$) (23, 24), two important prerequisites for a successful search for radiogenic ^{53}Cr from the decay of ^{53}Mn . Here, we report an ion microprobe study of the ^{53}Mn - ^{53}Cr isotope systematics of fayalite in type I chondrules in a Bali-like oxidized CV3 chondrite, Mokoia, to determine the time scale of alteration and thus the environment in which alteration of the CV chondrites occurred.

Type I chondrules in Mokoia are composed mainly of forsteritic olivine (Fa $_1$) and low-Ca pyroxene (Fs $_1$ Wo $_1$) phenocrysts, interstitial

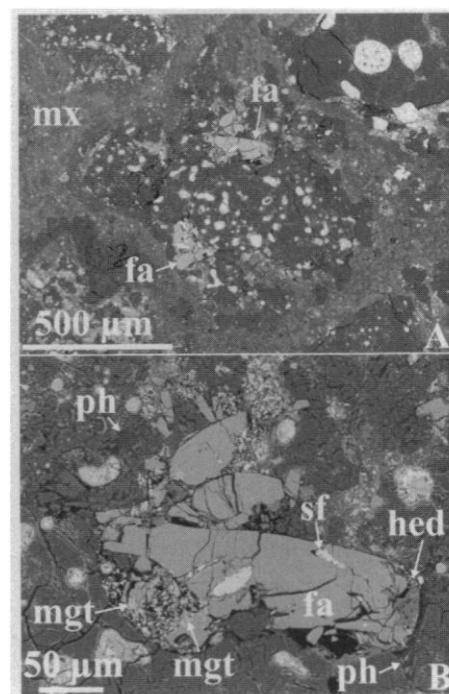


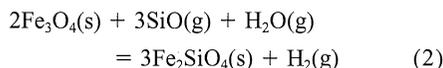
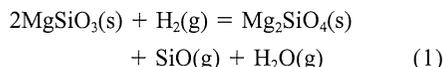
Fig. 1. Backscattered electron images of the oxidized CV3 chondrite Mokoia. (A) Irregularly shaped, type I (Mg-rich) chondrule 8 in the center is surrounded by a fine-grained matrix (mx) and contains numerous nodules of magnetite, sulfides, and associated fayalite (Fa $_{>96}$) surrounded by darker olivine (Fa $_1$) and pyroxene (Fs $_1$ Wo $_1$). (B) The upper fayalite grain in (A), showing inclusions of magnetite (mgt) and sulfide (sf). The fayalite appears to have been replaced by hedenbergite (hed), which is in direct contact with phyllosilicate (ph). This fayalite shows the largest excesses of ^{53}Cr (see Fig. 2).

I. D. Hutcheon and D. L. Phinney, Lawrence Livermore National Laboratory (LLNL), Livermore, CA 94551, USA. A. N. Krot, K. Keil, E. R. D. Scott, Hawai'i Institute of Geophysics and Planetology, School of Ocean and Earth Science and Technology, University of Hawai'i at Manoa, Honolulu, HI 96822, USA.

*To whom correspondence should be addressed.

anorthitic mesostases with crystals of high-Ca pyroxene (Fs₁Wo₃₅₋₄₅), and abundant magnetite-sulfide nodules (Fig. 1A). Chondrule mesostases and pyroxenes are replaced by the phyllosilicates, saponite, and Na-phlogopite (Table 1) (14). Fayalite (Fa₉₈₋₁₀₀) is associated with magnetite, sulfides, and hedenbergitic pyroxenes (~Fs₄₅Wo₅₀) (Fig. 1B) and contains ~0.2 to 0.9 weight % MnO and <0.02 weight % Cr₂O₃ (Table 1). Fayalite grains typically contain small inclusions of pure magnetite (Fe₃O₄) (Fig. 1B). Type I chondrules contain two types of magnetite: (i) grains in rounded nodules containing inclusions of sulfides and other phases and high abundances of minor elements (up to 2 weight % of Cr, P, Ca, and Mg), and (ii) massive magnetite associated with these nodules, containing fewer inclusions and much lower abundances (<0.02 weight %) of minor elements. Fayalite preferentially replaces massive magnetite (24, 25).

Two models have been proposed to explain the origin of the fayalite. According to the nebular model (24), fayalite was produced by high-temperature (>800°C) reactions between SiO gas, released by decomposition of pyroxene, and magnetite:



According to the asteroidal model, fayalite formed at low temperatures (<300°C) during fluid-rock interactions (5, 12, 25):

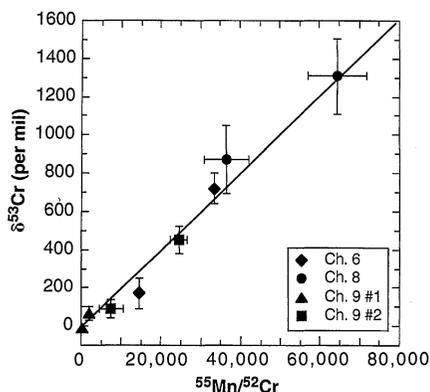
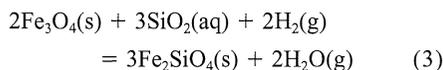


Fig. 2. ⁵³Mn-⁵³Cr evolution diagram for four fayalite grains in three magnesian (type I), porphyritic olivine-pyroxene chondrules (Ch. 6, 8, and 9) in the oxidized CV3 chondrite Mokoia. The slope of the line fitted to the data yields the initial ⁵³Mn abundance at the time of fayalite formation, (⁵³Mn/⁵⁵Mn)₀ = 2.32 (±0.18) × 10⁻⁶. The data are plotted as δ⁵³Cr, the per mil deviation from the ⁵³Cr/⁵²Cr ratio measured in a fayalite standard (26); errors are two standard deviations of the mean.

The Cr isotope compositions of four fayalite crystals in three Mokoia chondrules (chondrules 6, 8, and 9) were determined with an ion microprobe (26). These fayalites have low Cr concentrations; only grain 1 from chondrule 9 contains >0.5 μg of Cr₂O₃ per gram of crystal, and fayalite 1 from chondrule 8 contains <50 ng of Cr₂O₃ per gram. Each crystal was analyzed in two different spots, each ~10 μm in diameter. Large variations in Mn/Cr ratios between the two spots of an individual crystal, and within an individual analysis on a single spot, reflect heterogeneity in the Cr₂O₃ content of fayalite of up to a factor of ~2 (Table 2 and Fig. 2). The uncertainties in Mn/Cr ratios, particularly for fayalite in chondrule 8, reflect this inhomogeneity in Cr₂O₃. All four Mokoia fayalites show well-resolved excesses of ⁵³Cr, ranging up to ~1300 per mil, correlated with the respective ⁵⁵Mn/⁵²Cr ratios (Table 2 and Fig. 2). The linear correlation between the magnitudes of the ⁵³Cr excesses and the respective Mn/Cr ratios is characteristic of the in situ decay of ⁵³Mn and demonstrates that these fayalite crystals formed while ⁵³Mn was still extant. The slope of the correlation line fitted to the data (Fig. 2), passing through the normal Cr isotope composition (δ⁵³Cr =

0) at Mn/Cr = 0, indicates an initial abundance of ⁵³Mn corresponding to ⁵³Mn/⁵⁵Mn = 2.32 (±0.18) × 10⁻⁶ at the time the fayalites formed and cooled below the closure temperature for Cr diffusion.

These data provide evidence for radiogenic ⁵³Cr in fayalite in a chondritic meteorite. A previous study of fayalite grains in the Kaba CV3 chondrite failed to detect excess ⁵³Cr and set an upper limit to the ⁵³Mn/⁵⁵Mn ratio of 7 × 10⁻⁶ (23), consistent with the lower value reported here. Figure 2 is equivalent to a "whole-rock" isochron in which the four fayalite crystals may be considered to be cogenetic, if the data lie within analytical uncertainties of the best-fit line and the fayalite grains can be assumed to have formed with a common initial ⁵³Cr/⁵²Cr ratio. Seven of eight analyses fall within two standard deviations of the correlation line, suggesting that these fayalite grains in the three Mokoia chondrules represent a cogenetic system and that the slope of the "isochron" can be used to infer the time of formation.

The use of ⁵³Mn as a chronometer requires Cr isotope differences between samples to be determined by differences in age and not by nebular isotope heterogeneity. The distribution of ⁵³Mn in the early solar nebula appears to

Table 1. Electron microprobe analyses of fayalite (fa), hedenbergite (hed), saponite (sap), and phlogopite (phl) in Mokoia chondrules. Uncertainties (in parentheses) are one standard deviation of the mean.

Oxide (weight %)	ch6		ch8			ch9
	fa	fa	hed	sap	phl	fa
SiO ₂	28.9 (0.1)	29.0 (0.1)	46.7	40.2	35.4	28.8 (0.3)
TiO ₂	<0.03	<0.03	0.06	0.05	0.04	<0.03
Al ₂ O ₃	<0.03	<0.03	<0.03	9.1	24.8	<0.03
Cr ₂ O ₃	<0.03	<0.03	<0.03	0.66	0.04	<0.03
FeO	68.7 (0.6)	69.5 (0.6)	27.4	3.2	4.2	69.7 (0.2)
MnO	0.54 (0.34)	0.60 (0.25)	0.34	<0.03	0.12	0.45 (0.10)
MgO	0.07 (0.06)	0.36 (0.19)	0.79	17.8	18.1	0.05 (0.04)
CaO	<0.04	<0.04	22.4	0.11	0.30	<0.04
Na ₂ O	<0.04	<0.02	<0.04	1.7	1.4	<0.04
K ₂ O	<0.04	<0.04	<0.04	0.33	0.29	<0.04
Total	99.3 (0.3)	99.4 (0.2)	97.7	73.2	84.7	99.0 (0.2)
Fa	99.8 (0.2)	99.1 (0.5)	-	-	-	99.9 (0.1)
Fs	-	-	47.6	-	-	-
Wo	-	-	49.9	-	-	-

Table 2. Cr isotope composition of Mokoia fayalite. Letters "a" and "b" designate two sections of an analysis on an individual spot, separated by large variations in Mn/Cr [see (26)]. Uncertainties are two standard deviations of the mean.

Fayalite	δ ⁵³ Cr (per mil)	⁵⁵ Mn/ ⁵² Cr
Chondrule 6, 1a	166 ± 75	14,800 ± 400
Chondrule 6, 1b	725 ± 80	33,200 ± 1200
Chondrule 8, 1a	848 ± 140	37,400 ± 5500
Chondrule 8, 1b	1307 ± 200	64,500 ± 7400
Chondrule 9, 1a	-9 ± 11	200 ± 25
Chondrule 9, 1b	68 ± 33	2125 ± 115
Chondrule 9, 2a	90 ± 44	7400 ± 2000
Chondrule 9, 2b	445 ± 75	24,800 ± 2200
Matrix	0.8 ± 1.0	0.5

have been heterogeneous on a solar system-wide scale but homogeneous within the region in which the chondritic meteorites originated (27). However, the solar system initial abundance of ^{53}Mn is not well determined, with inferred $^{53}\text{Mn}/^{55}\text{Mn}$ ratios spanning the range from 8.4×10^{-6} to 4.4×10^{-5} (27, 28). If we assume that the initial $^{53}\text{Mn}/^{55}\text{Mn}$ ratio appropriate to the chondrites is represented by the average value measured in several Allende CAIs, $4.4 (\pm 1.1) \times 10^{-5}$ (28), we would conclude that the Mokoia fayalites formed between 14 and 17 My after Allende CAIs. If, instead, we assume an initial $^{53}\text{Mn}/^{55}\text{Mn}$ ratio of 8.4×10^{-6} , inferred from an intercomparison of the Mn-Cr and Pb-Pb chronologies of CAIs and the angrite differentiated meteorites (21, 27), we would calculate an interval roughly half as long, ~ 7 My.

Observations of pre-main-sequence stars and chronological data of primitive meteorites have been used to infer a lifetime for the solar nebula of ~ 10 My, albeit with large uncertainty (29). The longer of the two Mn-Cr time scales places the formation of the Mokoia fayalites at least 4 My after the nebula is believed to have dissipated, which suggests that the Mokoia fayalites formed in an asteroid and not in the nebula. The shorter interval falls near the upper limit of nebular lifetimes but allows for the possibility of alteration in a nebular setting. However, independent of the Mn-Cr time scale, the inferred $^{53}\text{Mn}/^{55}\text{Mn}$ value in the Mokoia fayalites falls within the range of values determined for carbonates in CI and CM meteorites, believed to be produced by aqueous activity on asteroids (19, 20), and for several classes of differentiated meteorites (21). Because the initial $^{53}\text{Mn}/^{55}\text{Mn}$ ratio found here agrees with values inferred for meteorites formed in asteroids, we conclude that fayalite in the CV chondrites formed in an asteroid and not in the nebula. Because fayalite grains are partially replaced by Ca-Fe-rich pyroxenes (Fig. 1), we infer that Ca-Fe-rich minerals also formed in an asteroidal setting.

Our data indicate that although chondritic meteorites were affected by many different types of alteration processes, aqueous activity was occurring nearly simultaneously on the parent bodies of at least three classes of unequilibrated chondritic meteorites—CI, CM and CV—shortly after the parent bodies accreted. The time scale is consistent with heating of asteroids by ^{26}Al decay (30), leading to the production of liquid water by melting of ice, and suggests that water played an important role in the chemical evolution of primitive solar system bodies. This ^{53}Mn - ^{53}Cr chronology of early solar system events, developed from a diverse suite of samples including primitive chondrites and differentiated meteorites, leads to a scenario in which igneous activity and differentiation on some asteroidal bodies was

occurring contemporaneously with aqueous activity on other, unmelted asteroids shortly after CAIs and chondrules formed in the solar nebula and planetesimals accreted.

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- The Cr isotope analyses were performed with a modified Cameca ims-3F ion microprobe, using a 14.5-keV, $^{16}\text{O}^-$ primary ion beam with 3 to 5 nA current focused into a spot ~ 10 μm in diameter. Low-energy (0 to ~ 40 eV) positive secondary ions were accelerated nominally to 4500 V and mass analyzed at a mass resolving power of 3200 ($m/\Delta m$) to separate potential interfering species (for example, $^{24}\text{Mg}^{28}\text{Si}^+$, $^{25}\text{Mg}^{28}\text{Si}^+$) from the atomic Cr ions at masses 52 and 53. $^{52}\text{CrH}^+$ is not resolved at this mass resolving power but was determined to contribute $<0.1\%$ of the measured intensity at mass 53 by analyses of a synthetic fayalite standard at a mass resolving power of 6500. To minimize the ion signal arising from cracks filled with relatively Cr-rich matrix, we inserted a field aperture (effective diameter, 30 μm) into the sample image plane. Secondary ion intensities were corrected for background (0.015 s^{-1}) and dead time (17 ns); because of the very low Cr^+ intensity in Mokoia fayalites, only the background correction is significant and was always $<2.5\%$ of the ^{53}Cr intensity. Each analysis consisted of 10 to 20 blocks of 10 cycles each, with a cycle representing one pass through the masses ^{47}Ti , ^{49}Ti , ^{50}Cr , ^{52}Cr , ^{53}Cr , and ^{55}Mn . In the Mokoia fayalites, the intensity of $^{50}\text{Ti}^+$ was usually comparable to or greater than the $^{50}\text{Cr}^+$ intensity, and attempts to use the $^{50}\text{Cr}/^{52}\text{Cr}$ ratio (after correcting for ^{50}Ti overlap) to determine the instrumental mass fractionation proved imprecise and inaccurate. Excesses of ^{53}Cr were instead determined relative to the mean $^{53}\text{Cr}/^{52}\text{Cr}$ ratio measured in a synthetic fayalite and are reported as $\delta^{53}\text{Cr}$, the deviation (per mil) from the fayalite reference value. At least two analyses of the fayalite standard were performed before analyzing each Mokoia chondrule. Over the course of this study, 14 analyses of the standard yielded a mean $^{53}\text{Cr}/^{52}\text{Cr}$ ratio value of 0.11262 ± 0.00026 , indicating an instrumental mass fractionation of 6.8 per mil favoring the lighter Cr isotopes, relative to the NIST standard value, $^{53}\text{Cr}/^{52}\text{Cr} = 0.113389$ (31). Uncertainties in the $\delta^{53}\text{Cr}$ values are twice the standard deviation of the block averages and include the reproducibility of the fayalite standard and the internal measurement precision on an individual analysis; because of the low Cr contents, the uncertainties reflect primarily counting statistics on ^{53}Cr . The conductive carbon coat on the Mokoia section contained a higher Cr concentration than the fayalites; the first two blocks of data were excluded from all analyses to avoid contamination by this extraneous Cr. Although we were able to reduce contamination, complete removal of the surficial Cr proved unfeasible. Each analysis of Mokoia fayalite was split in two, listed as "a" and "b" in Table 2, with the "a" section always having higher Cr^+ intensity and comparably lower Mn/Cr ratios. The Mn/Cr atomic ratios were calculated from measured $^{55}\text{Mn}^+ / ^{52}\text{Cr}^+$ ion ratios using a sensitivity factor of 1.17 ± 0.05 , determined from analyses of San Carlos olivine in which the Cr content had been determined by instrumental neutron activation analysis.
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