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Hf–W, Sm–Nd, and Rb–Sr isotopic evidence of late impact fractionation and mixing of silicates on iron meteorite parent bodies

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Abstract

We report the first Sm–Nd and Rb–Sr isotopic analyses of silicate inclusions in four IIE iron meteorites: Miles, Weekeroo Station A and B, and Watson. We also report the Hf–W isotopic composition of a silicate inclusion from Watson and $^{182}W/^{184}W$ of the host FeNi metal in all four IIEs. The host metal in Watson has a negative ε_W value (-2.21 ± 0.24), similar to or higher than other iron meteorites [1,35] and consistent with segregation of metal from silicate early in solar system history. However, the large silicate inclusion in the Watson IIE iron yielded a chondritic ε_W value (-0.50 ± 0.55), thus indicating a lack of equilibration with the FeNi host within the practical lifetime of activity of the parent ^{182}Hf (~ 50 Ma). One of the silicate inclusions in Miles is roughly chondritic in major-element composition, has a present-day ε_{Nd} of +10.3, relatively non-radiogenic $^{87}Sr/^{86}Sr$ (0.714177 ± 13), and a T_{CHUR} age of 4270 Ma. Two silicate inclusions from Weekeroo Station and one from Watson exhibit fractionated Sm/Nd and Rb/Sr ratios, and more radiogenic $^{87}Sr/^{86}Sr$ (0.731639 ± 12 to 0.791852 ± 11) and non-radiogenic ε_{Nd} values (-5.9 to -13.4). The silicate inclusion in Watson has a T_{CHUR} age of 3040 Ma, in agreement with previously determined ^4He and ^{40}Ar gas retention ages, indicative of a late thermal event. A later event is implied for the two silicate inclusions in Weekeroo Station, which yield indistinguishable T_{CHUR} ages of 698 and 705 Ma. Silicate inclusions in IIE iron meteorites formed over a period of 3 billion yr by impacts, involving an H-chondrite parent body and an FeNi metal parent body. The LILE-enriched nature of some of these silicates suggests several stages of melting, mixing, and processing. However, there is little evidence to suggest that the silicates in the IIE irons were ever in equilibrium with the host FeNi metal.

Keywords: iron meteorites; isotopes; FeNi metal; chronology; impact; silicate inclusion

1. Introduction

It was long thought that iron meteorites repre-

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sent the cores of asteroids that have undergone extensive melting and separation from silicates. These irons were often split into groups based upon whether they were thought to have undergone fractional crystallization (magmatic irons) versus those which showed no such evidence (non-magmatic) [45,49]. However, more recent work has allowed a different or modified interpretation for several groups of iron meteorites, especially those that contain enclosed silicates (e.g., [29]).

The age of the silicates contained in some irons has been vigorously pursued. Over 30 yr ago, Bogard et al. [2], Burnett and Wasserburg [3,4], and Sanz et al. [5] used inclusions of silicate material in iron meteorites to date, presumably, the age of the iron. Although some of the silicate inclusions in certain irons yielded Rb–Sr ages indistinguishable from earliest solar system materials (e.g., Colomera, 4.61 ± 0.04 Ga), others indicated formation events distinctly younger (e.g., Kodaikanal, 3.8 ± 0.1 Ga). These younger ages were taken to represent not only the age of the silicate inclusion, but the age of the host FeNi metal [3]. However, no independent test has been undertaken to simultaneously date the silicate inclusion and the host FeNi metal in an iron meteorite. Recent advances in analysis of the short-lived nuclide ^{182}Hf allow just such an age comparison to be made.

Although other groups of irons (namely IAB and IIICD; [49]) contain silicate inclusions, the IIE group of iron meteorites routinely contains abundant silicate inclusions, often making up over 20% of the sample [6]. Based upon trace-element abundances, the IIE irons have been subdivided into two groups – those that exhibit major- and trace-element abundances and patterns similar to H-group chondrites, termed the unfractionated group, and those that exhibit abundance patterns that are fractionated relative to H-group chondrites. Three models have been put forth for the origin of silicate inclusions in IIE irons: (1) a near-surface model involving mixing of IIE metal with H-chondrite silicate melts [7,8]; (2) mixing of metallic and silicate melts at the core–mantle boundary in a chondritic parent body [9], and (3) a combination of impact melting and planetary differentiation [6].

To test these hypotheses, we have performed a radiogenic isotopic study of four silicate inclusions in three different IIE iron meteorites – two in Weekeroo Station, and one each in Watson and Miles. Furthermore, to constrain the evolution of these silicate inclusions, as well as their host FeNi metals, we have examined both short-lived (Hf–W) and long-lived (Rb–Sr, Sm–Nd) isotopic systems. Both W and Hf are refractory and found in chondritic proportions in planetoids. However, W is moderately siderophile and Hf is strongly lithophile, thus, metal–silicate fractionation leads to partitioning of W into FeNi metal and Hf into silicate. If this metal–silicate fractionation occurs within 50 Ma of planetary evolution, then ε_W anomalies (positive in the silicate, negative in the FeNi metal) should result [1,46]. Indeed, carbonaceous chondrites have ε_W values (= deviation of ^{182}W from chondritic values at any given time) that are roughly zero, HEDs (howardites, eucrites, and diogenites, thought to come from the eucrite parent body; i.e., asteroid 4-Vesta) and some SNC (shergottites, nakhlites, and chassignites; thought to come from Mars) meteorites have ε_W values that are distinctly positive, and metals from both iron meteorites and ordinary chondrites have ε_W values that are distinctly negative [1,10].

2. Previous isotopic studies of metal and silicate in iron meteorites

Stewart et al. [11] were the first to analyze Sm–Nd isotopes in a silicate inclusion in an iron meteorite (Caddo County, IAB), but no silicate inclusions in IIE iron meteorites have been studied. The Caddo Co. silicate exhibited a roughly chondritic initial ^{143}Nd/^{144}Nd isotopic composition ($\varepsilon_{143} = -0.1 \pm 0.2$) and a well-defined ^{147}Sm/^{144}Nd age of 4.53 ± 0.02 Ga. A plagioclase separate from this sample yielded the lowest measured ^{142}Nd/^{144}Nd to date ($\varepsilon_{142} = -2.4 \pm 0.4$), consistent with early silicate fractionation in the IAB parent body [11]. Based on cooling rates and their Sm–Nd studies, Stewart et al. [11] preferred a model of formation for the IAB Caddo Co. silicates by incorporation within the FeNi metal host at depths > 2 km, early (within first 30 Myr) during parent body differentiation.

2.1. Isotopic studies and ages for IIE silicate inclusions

Previous isotopic studies of silicates in IIE irons have indicated a diversity of ages which led Olsen et al. [12] to subdivide the IIEs into two groups. A young group (3.7 ± 0.2 Ga) contains the irons Watson, Kodaikanal, and Netschaevo, whereas an older group (4.5 ± 0.1 Ga) is composed of Colomera, Miles, Techado, and Weekeroo Station. This subdivision crosses previously determined mineral–chemical boundaries, as both age groups include samples from the fractionated and unfractionated groups. In the discussion below, we have recalculated the Rb–Sr ages and ^{87}Sr/^{86}Sr initial ratios for analyzed silicate inclusions in Kodaikanal [3] and Colomera [5] using a ^{87}Rb decay constant of 1.402×10^{-11} yr^{-1} [13] and the program ISOPLOT [14]. This decay constant for ^{87}Rb is based on the convergence of ages for Rb–Sr whole-rock data and U–Th–Pb data from chondritic meteorites [13].

2.1.1. Young group

The fractionated silicates in the Kodaikanal IIE iron have been studied intensively. Bogard et al. [15] determined K–Ar ages of 3.3 ± 0.1 and 3.5 ± 0.1 Ga (K–Ar) for silicates in Kodaikanal. Burnett and Wasserburg [4] divided the silicate from Kodaikanal into 11 density and mineral separates. All 11 separates from Kodaikanal yield a well-defined line and an age of 3743 ± 44 Ma (MSWD = 1.58) with a poorly defined initial ^{87}Sr/^{86}Sr (due to large errors in the analyses) of 0.719 ± 0.011. Gopel et al. [16] also obtained a concordant U–Pb age (3676 ± 3 Ma) for Kodaikanal which is similar to this Rb–Sr age. Most significantly, silicate inclusions from Kodaikanal point to a relatively young age (3.67–3.75 Ga), possibly up to 800 Myr after formation of the host FeNi metal.

The unfractionated Watson meteorite has yielded a K–Ar age of 3.5 ± 0.2 Ga [12] and, recently, a more precise ^{40}Ar–^{39}Ar age of 3.656 ± 0.005 Ga [17]. An ^{40}Ar–^{39}Ar analysis of the silicate from the unfractionated group meteorite Netschaevo is the only other IIE that has yielded a similarly young age (3.79 ± 0.03 Ga; [18]).

2.1.2. Old group

The unfractionated Techado IIE iron has been dated by both K–Ar and ^{40}Ar–^{39}Ar methods. The K–Ar determination yielded only a minimum age of 4.2 Ga [8], but the ^{40}Ar–^{39}Ar method defined an age of 4.482 ± 0.025 Ga [17]. Garrison and Bogard [17] have also determined an ^{40}Ar–^{39}Ar age for the fractionated Miles meteorite of 4.408 ± 0.011 Ga.

Sanz et al. [5] separated silicate inclusions from the fractionated Colomera IIE iron into 14 mineral separates, but did not include the three pyroxene points or a single anorthite point in their regression. Once the most radiogenic 'outer' separates are included with the previous data for Colomera, making a total of 20 separates, the age is increased to 4590 ± 127 Ma (MSWD = 65.5), albeit with the same initial ^{87}Sr/^{86}Sr (0.6994 ± 0.0004). However, the high MSWD values for both Colomera silicate ages indicate that the scatter is not solely due to analytical error. Regardless of possible problems with the Rb–Sr analyses for Colomera, there is still a suggestion of a silicate age which may be commensurate with the age of the FeNi metal.

Previous isotopic studies of silicate inclusions in the Weekeroo Station IIE iron included Rb–Sr and K–Ar determinations. Burnett and Wasserburg [4] analyzed size and density separates from a silicate inclusion which yielded a Rb–Sr 'age' of 4.33+0.23/−0.12 Ga and an initial ^{87}Sr/^{86}Sr of 0.703. Evensen et al. [19] analyzed four separate inclusions which yielded a well-defined Rb–Sr whole-rock isochron of 4.39 ± 0.07 Ga and an initial ^{87}Sr/^{86}Sr of 0.7013. Bogard et al. [2] also analyzed a group of density and size separates of a silicate inclusion from Weekeroo Station for K–Ar. An average of four analyses yielded an age of 4.49 ± 0.10 Ga. All of these ages overlap, within analytical uncertainty. Furthermore, it would appear that the silicate inclusions in Weekeroo Station, although likely coeval [29], may exhibit a range in initial $^{87}Sr/^{86}Sr$ ratios (0.7013–0.703).

2.2. Re–Os isotopic studies

With the advent of the ^{187}Re–^{187}Os isotopic system and its widespread use in planetary geochemistry, several groups have analyzed the FeNi metal in several classes of iron meteorites to determine their ages. Earlier studies assumed that iron meteorites were coeval with chondrites, used their colinear relationship on a $^{187}Re/^{186}Os$ versus $^{187}Os/^{186}Os$ diagram, and assumed an age of 4550 Ma to calculate the decay constant for ^{187}Re ($1.53 \pm 0.08 \times 10^{-11}$ yr^{-1}; although originally it was reported as 1.62×10^{-11} yr^{-1}) [20,21]. However, Lindner et al. [22] determined a much larger decay constant (shorter half-life) for ^{187}Re ($1.639 \pm 0.050 \times 10^{-11}$ yr^{-1}), leading to the suggestion that iron meteorites might be distinctly younger than chondrites. Subsequently, Morgan et al. [23], Horan et al. [24], and Morgan et al. [25] analyzed several samples from groups IA, IIA, IIAB, IIB, IIIA, and IIIAB and confirmed that iron meteorites are generally similar in age to chondrites, yielding isochrons of 4.58 ± 0.04 Ga (IIAB) and 4.55 ± 0.11 Ga (IIIA). Smoliar et al. [47] did the most precise and complete study to that time, calculating ages for the IIA, IIIA, IVA, and IVB irons of 4.537 ± 8, 4.558 ± 12, 4.464 ± 26, and 4.527 ± 29 Ga, respectively. They also redefined the ^{187}Re decay constant to 1.666×10^{-11} yr^{-1} based on convergence of the Pb–Pb ages for angrites with the IIIA Re–Os isochron [47]. Shen et al. [26] analyzed four IIAB, four IVA, three IVB, three IAB, and two IIIAB iron meteorites for Re–Os isotopes and generated a well-defined isochron which defines a possible 'mixed' age of 4.62 ± 0.02 Ga. Samples within particular groups gave similar ages, although there was a suggestion of a 60 ± 45 Ma time difference between 'older' IVA irons (4.67 ± 0.04 Ga) and 'younger' IIAB irons (4.61 ± 0.01 Ga). However, Smoliar et al. [27,47] have indicated somewhat younger ages for groups IIA (4.537 ± 0.008 Ga) and IIB (4.43 ± 0.05 Ga). Birck and Allegre [28] determined a well-defined isochron of 4.624 ± 0.017 Ga for a group of iron meteorites, including those from groups IA, IIA, IVB, and IIE. They found that several FeNi metal samples in Kodaikanal (IIE), the iron which yielded U–Pb and Rb–Sr ages of 3.67–3.75 Ga in the silicate (see above), were concordant with the other old FeNi metal ages [28].

Re–Os isotopic systematics of certain iron meteorites also have indicated possible late disturbances on the parent bodies and/or later magmatic events. A group of IIB irons (the Navajo subgroup) contained excess ^{187}Os that was interpreted to be due to either (1) crystallization from an evolved portion of the asteroid that had remained molten for some exceedingly lengthy time after most IIABs, or (2) impact remelting of the core hundreds of millions of years after its initial crystallization, or (3) Os diffusion into the Navajo group irons several hundred million years after crystallization (at 3.1–3.5 Ga). Shen et al. [26] analyzed schreibersites from group IAB and IIAB irons which also contained excess radiogenic ^{187}Os. They attributed these excesses to open-system behavior of the schreibersites up to 1 Ga after formation of the FeNi metal.

The present status of isotopic studies on iron meteorites and their enclosed silicate inclusions leaves several unresolved questions. Is the formation of asteroidal cores, as represented by the magmatic iron meteorites, restricted to the first 100 Myr, possibly the first 10–40 Myr [47] of the solar system [35]? If so, then how does one explain the young (3.8 Ga) Rb–Sr age for the silicate inclusions in the IIE iron Kodaikanal? Were the FeNi metal and silicate formed at different times in IIE irons? If so, what mechanisms can be proposed to 'inject' these silicates into previously crystallized FeNi metal? If silicate was injected into previously crystallized metal, what effect did this process have on the metal? Was the metal molten or solid at the moment of metal–silicate mixing?

3. Silicate petrography and mineral chemistry

In depth petrographic and mineral–chemical analyses of silicate inclusions in Weekeroo Station have been published in a companion paper [29]. Weekeroo Station samples A and B were excavated from the same slab of sample AMNH (American Museum of Natural History) 2620 [29]. Weekeroo Station A consists predominantly of 30% augite phenocrysts in a cryptocrystalline (glass; 59%) mesostasis which was formerly feldspar and silica. Weekeroo Station A also contains small proportions of orthopyroxene (5%) and pigeonite (3%) and trace amounts of (in decreasing abundance) phosphate, chromite, troilite, metal, and ilmenite. The silicate inclusion known as Weekeroo Station B consists of 56% feldspar, 13% augite with coarse exsolution of orthopyroxene, orthopyroxene grains (15%), and pigeonite (10%), in a matrix of silica (3%). Minor amounts of (in decreasing order) phosphate, troilite, metal, K-feldspar, and chromite are also present. The matrix in Weekeroo Station B is demonstrably coarser than in Weekeroo Station A.

A split of the silicate inclusion in Watson that we studied contains on average 57% olivine (10–100 μm) poikilitically enclosed by large (400–1000 μm) orthopyroxenes (23%), 12% feldspar, 5% augite (200–400 μm), and (in decreasing order) minor phosphate (mostly whitlockite), troilite, euhedral to anhedral chromite (200–350 μm), and FeNi metal [12]. Unlike many other IIE silicates, chondrules, corona structures, and ilmenite are absent.

Ikeda and Prinz [6] and Ikeda et al. [30] have studied a total of 49 separate inclusions from the Miles IIE iron, and they have subdivided the samples texturally into gabbroic and cryptocrystalline varieties. The gabbroic silicates generally have irregular contacts with the FeNi metal, whereas cryptocrystalline samples exhibit ellipsoidal, lobate, and rounded contacts with the enclosing FeNi metal. The contacts between the silicate inclusions and the FeNi metal are often defined by the presence of schreibersite. The single inclusion that we studied contained approximately 70% feldspar and 30% augite.

4. Analytical methods and data presentation

Silicate samples were crushed in an aluminum oxide mortar under a flow of better than class 100 air and then split into two portions, a smaller, Sm–Nd–Rb–Sr split, and a larger Hf–W split. The smaller split (roughly 50 mg) was then dissolved in HF, HNO_3, and HCl, and isotope dilution measurements made on a 10–15% split of this solution with ^{87}Rb–^{84}Sr and ^{149}Sm–^{150}Nd mixed spikes. Total-process blanks for chemical procedures were always less than 10 pg Rb, 120 pg Sr, 10 pg Sm, and 50 pg Nd. Sr and Nd isotopic data were obtained by multidynamic analysis on a VG Sector multicollector mass spectrometer. All Sr and Nd isotopic analyses are normalized to $^{86}\mathrm{Sr}/^{88}\mathrm{Sr} = 0.1194$ and $^{146}\mathrm{Nd}/^{144}\mathrm{Nd} = 0.7219$, respectively. Analyses of SRM 987 Sr and La Jolla Nd standards were performed throughout this study and gave weighted averages (at the 95% confidence limit, external precision) of $^{87}\mathrm{Sr}/^{86}\mathrm{Sr} = 0.710250 \pm 0.000011$, and $^{143}\mathrm{Nd}/^{144}\mathrm{Nd} = 0.511854 \pm 0.000011$, respectively. Internal, within-run, statistics are almost always of higher precision than the external errors (see Table 2 for within-run statistics of samples, which are comparable to that of the standards). All isotope dilution measurements utilized static mode multicollector analyses.

By convention, the Nd isotopic data are also presented in Tables 1 and 2 in epsilon units, deviation relative to a chondritic uniform reservoir, CHUR (DePaolo, 1976):

$$\varepsilon_{\mathrm{Nd}} = [(^{143}\mathrm{Nd}/^{144}\mathrm{Nd}_{\mathrm{sample}} - {}^{143}\mathrm{Nd}/^{144}\mathrm{Nd}_{\mathrm{CHUR}})/\ {}^{143}\mathrm{Nd}/^{144}\mathrm{Nd}_{\mathrm{CHUR}}] \times 10^4$$

Model ages (or single-stage evolution ages) have been calculated for Nd isotopes:

$$T_{\mathrm{CHUR}} = 1/\lambda \times \ln[(^{143}\mathrm{Nd}/^{144}\mathrm{Nd} - 0.512638)/\ (^{147}\mathrm{Sm}/^{144}\mathrm{Nd} - 0.1966) + 1]$$

The T_{CHUR} model age is determined relative to a present-day CHUR with $^{143}\mathrm{Nd}/^{144}\mathrm{Nd} = 0.512638$ and $^{147}\mathrm{Sm}/^{144}\mathrm{Nd} = 0.1966$. Errors in the model ages are estimated from consideration of errors in the parent–daughter ratios and measured isotopic ratios. Errors in isochron ages are 2-σ [31]

Table 1
Hf–W isotopic composition of metals and a silicate inclusion in IIE iron meteorites

Sample	AMNH#	Hf (ppb)	W (ppb)	^{180}Hf/^{184}W	^{182}W/^{184}W	ε_W
Watson	4762					
FeNi		0	1010	0	0.864809 ± 21	−2.21 ± 0.24
Silicate		154.2	59.8	3.040	0.864957 ± 48	−0.50 ± 0.55
Miles (a)	4866				0.864764 ± 19	−2.73 ± 0.22
Miles (b)	4866				0.864755 ± 31	−2.83 ± 0.36
Weekeroo (a)	2620				0.864737 ± 17	−3.04 ± 0.20
Weekeroo (b)	2620				0.864727 ± 17	−3.16 ± 0.20

of the scatter as calculated in the ISOPLOT program of Ludwig [14].

The larger of the two silicate splits was used for Hf and W abundance measurements and W isotopic composition analyses [10,32]. This split was leached with 1 M HCl to remove saw marks and fusion crust material. The samples were then digested sequentially with concentrated HF, 8 M HNO_3, and 6 M HCl. Roughly 10% of the solution was then separated and spiked with ^{178}Hf and ^{186}W and the remaining 90% of the solution was dried and redissolved in 8 ml of 4 M HF. The chemical separations were similar to those of Salters and Hart [33], although on a smaller scale, using columns filled with 3.5 ml of Bio-Rad AG1x8 (200–400 mesh) anion resin. The Hf and W were eluted and collected with a mixed solution of 6 M HCl+1 M HF. The same chemical separation procedure was used for the spiked solutions, except the column volume was 1 ml, and Hf and W were collected together. The total W procedural blank is ~0.4 ng, which is negligible for all but the Watson silicate sample. The W isotopic compositions were measured on a VG Plasma-54 multiple-collector inductively coupled plasma mass spectrometer (MC-ICPMS) [34]. Standards (NIST-3163) were routinely run between samples to monitor performance and W memory effects. Tungsten isotopic measurements were normalized to $^{186}\text{W}/^{184}\text{W} = 0.927633$. The concentrations of Hf and W were determined by isotope dilution on the MC-ICPMS. The deviation of ^{182}W/^{184} from chondritic is determined by the notation:

$$\varepsilon_W = \{[(^{182}\text{W}/^{184}\text{W})_{\text{sample}}/(^{182}\text{W}/^{184}\text{W})_{\text{NIST-3163}}]-1\}\times 10^4 \quad (1)$$

5. Hf–W isotopic compositions of IIE irons

The ε_W values for the FeNi metal samples in the IIEs Weekeroo Station (two splits) and Miles are about −3, at the high end of the spectrum of values obtained from iron meteorites [1,35]. With the exception of Goose Lake, the IIEs yield the least negative ε_W values among all iron meteorites (Fig. 3).

5.1. Isotopic composition of the silicate inclusion and host FeNi metal in Watson

In comparison with other iron meteorites [1,35],

Table 2
Rb–Sr, Sm–Nd isotopic composition of silicate inclusions in IIE iron meteorites

Sample	Rb (ppm)	Sr (ppm)	^{87}Rb/^{86}Sr	^{87}Sr/^{86}Sr	Sm (ppm)	Nd (ppm)	^{147}Sm/^{144}Nd	^{143}Nd/^{144}Nd	$\varepsilon_{Nd(0)}$	T_{CHUR} (Ma)
Miles-4	4.61	66.2	0.2005	0.714177 ± 13	0.731	2.05	0.2154	0.513168 ± 19	+10.3	4270
Watson	9.07	17.8	1.482	0.791852 ± 11	0.235	0.782	0.1815	0.512333 ± 23	−5.9	3040
Weekeroo-A	9.09	37.5	0.7004	0.752649 ± 11	0.649	8.45	0.04646	0.511950 ± 8	−13.4	698
Weekeroo-B	8.29	40.8	0.5861	0.731639 ± 12	0.737	5.96	0.07481	0.512075 ± 10	−11.0	705

the Watson metal has a less negative ε_W anomaly (-2.21 ± 0.24), indicating later segregation or equilibration with silicate early in solar system evolution (Table 1). A >1 g sample of a large silicate inclusion from the Watson IIE iron exhibits a chondritic ε_W value (-0.50 ± 0.55), despite a high, non-chondritic ^{180}Hf/^{184}W (3.040; compared to chondritic, which is 1.57 [1]), and indicating late derivation from chondritic materials and/or reequilibration with chondritic materials, in contrast to the W isotopic composition of the host FeNi metal.

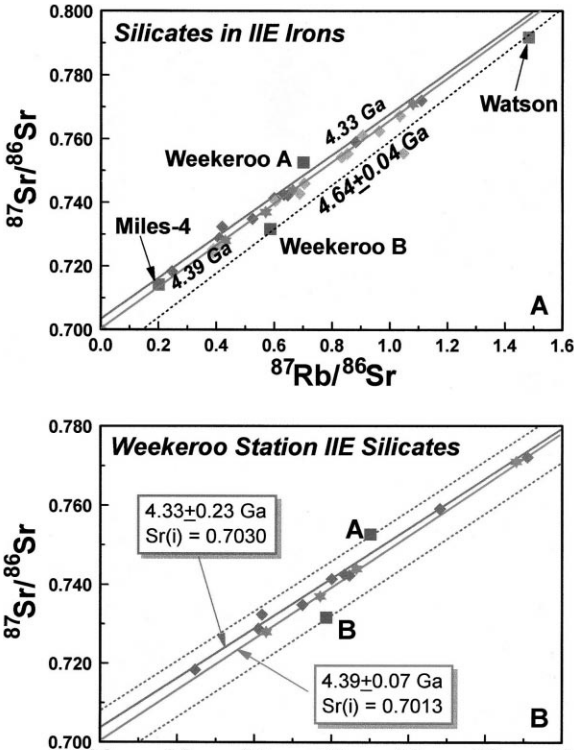

Fig. 1. Measured ^{87}Rb/^{86}Sr versus ^{87}Sr/^{86}Sr for IIE silicates. (A) Plot of bulk analyses of silicates in IIE irons. A regression of Watson and Weekeroo Station B yields an 'age' of 4.64 ± 0.04 Ga. Reference isochrons at 4.33 Ga and 4.39 Ga are also shown which encompass all of the data, except those for Weekeroo Station A and B and Watson. (B) Plot of bulk silicate and silicate separates from Weekeroo Station (diamonds = [3,4]; stars = [19] – estimated from a figure in their abstract; and this study, Table 2). A regression of the Evensen et al. [19] points yields an age of 4.35 ± 0.07 Ga and that for the Burnett and Wasserburg [3,4] data gives an age of 4.33 ± 0.23 Ga (using $\lambda_{\mathrm{Rb}} = 1.402 \times 10^{-11}$).

6. Rb–Sr, Sm–Nd isotopic compositions of silicate inclusions

All four of the silicate inclusions analyzed are notably enriched in Rb, although to varying degrees (Table 2). The silicate analyzed in Miles (designated Miles-4) has the lowest Rb/Sr (Fig. 1A) and is otherwise relatively depleted in the LILE, exhibiting a Sm/Nd ratio above chondritic and a positive $\varepsilon_{\mathrm{Nd}}$ (+10.3; Table 2; Fig. 2). This supra-chondritic Sm/Nd ratio (i.e., LREE depletion) is consistent with ion probe trace-element data on augite (containing the highest REE abundances among the major minerals, thus controlling the bulk REE pattern) from IIE silicate inclusions in Colomera [36] and Weekeroo Station [29]. The old T_{CHUR} age (4270 Ma) suggests that this silicate has undergone little, if any, post-crystallization modification. However, the ^{147}Sm/^{144}Nd ratios for both Miles-4 and Watson differ from chondritic values by only about 9% and 8%, respectively. Thus, calculations of model T_{CHUR} values are relatively imprecise for these samples.

The two inclusions studied from Weekeroo Station (A and B) yield similar results, are both extremely LREE-enriched (i.e., very low Sm/Nd), and exhibit negative $\varepsilon_{\mathrm{Nd}}$ values (-11.0 to -13.4). Assuming derivation from a chondritic source, the event which caused this enrichment must have occurred during the last 1 Gyr (Fig. 2), as T_{CHUR} ages are indistinguishable at 698–705 Ma. Rb–Sr isotopic determinations for these two samples were similarly enriched in Rb and are as radiogenic as those reported by Burnett and Wasserburg [3,4], although our samples do not lie along their 4.37 Ga (recalculated to 4.33 Ga using $\lambda_{\mathrm{Rb}} = 1.402 \times 10^{-11}$ yr^{-1}) isochron (Fig. 1B).

The silicate in Watson is among the most Rb-enriched and most radiogenic of bulk silicate inclusions measured in IIE irons (Fig. 1A). Burnett and Wasserburg [3,4] did find inclusions (D5 and D6) in the Colomera and Kodaikanal IIE irons which have higher Rb/Sr and, consequently, are more radiogenic (^{87}Sr/^{86}Sr up to 2.052). Due to

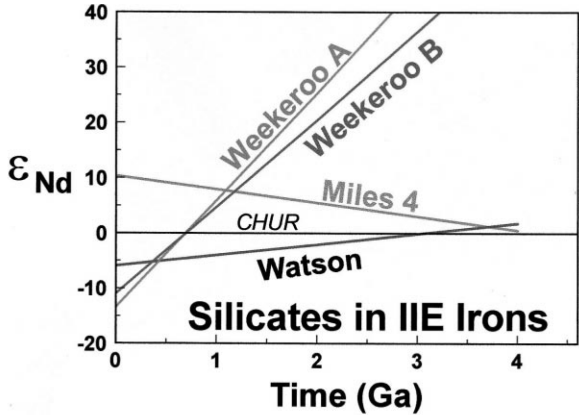

Fig. 2. Nd isotopic evolution diagram showing the single-stage evolution of radiogenic Nd ($\varepsilon_{Nd} = [(^{143}Nd/^{144}Nd_{sample} - ^{143}Nd/^{144}Nd_{CHUR})/^{143}Nd/^{144}Nd_{CHUR}] \times 10^4$) in IIE iron meteorite silicate inclusions over time. Note that the two silicates from Weekeroo Station converge at the CHUR at 700 Ma. The silicate inclusion in Miles crosses the CHUR evolution line at 4270 Ma, and that for Watson crosses CHUR at 3040 Ma.

their elevated Rb/Sr, mineral and density separates in Kodaikanal exhibited even more radiogenic isotopic compositions; up to $^{87}Sr/^{86}Sr$ values of 8.847 for 'feldspar glass' [3,4].

The Sm–Nd systematics of the Watson silicate ($\varepsilon_{Nd} = -5.9$) appear to be intermediate between that of Miles-4 and Weekeroo Station, which is inconsistent with its relatively elevated Rb and radiogenic $^{87}Sr/^{86}Sr$. The Watson silicate yields a relatively imprecise T_{CHUR} of 3040 Ma (Fig. 2).

7. Discussion

7.1. Hf–W in silicate inclusions, FeNi metal, and lack of equilibration

Lee and Halliday [1] found Arispe to be the least radiogenic iron meteorite and concluded that Arispe was the first of the irons to undergo silicate–metal fractionation. The ages of silicate–metal fractionation were then referenced to this earliest fractionation in Arispe. Using this logic, the IIE irons would appear to have formed later than most (if not all) other iron meteorites, or > 8 Ma after Arispe (Fig. 3). Conversely, the more radiogenic character of the IIE irons could be due to an admixture of material that did not experience early metal–silicate fractionation, such as bulk chondrite material, or an admixture of material that did fractionate, but had a positive ε_W.

If the silicate inclusion in Watson was cogenetic with the metal in Watson, as has been postulated for other magmatic iron meteorites [35], then it should exhibit a complementary positive ε_W isotopic anomaly. Positive ε_W anomalies are observed for many of the silicates derived from asteroids and planets, such as some SNC and HED achondrites (Fig. 4) [10]. Such positive ε_W values are consistent with core formation early in solar system history creating a high Hf/W in the resulting mantle. However, a positive ε_W anomaly is not present in the Watson silicate; therefore, the silicate and metal do not appear to be cogenetic. Metal–silicate mixing for Watson must have occurred later, after ^{182}Hf had largely decayed. The data can be explained if metal in Watson was derived from a source region that experienced early metal–silicate fractionation, and if silicate in Watson was derived from a different source region that did not undergo metal–silicate fractionation. One reasonable explanation for this is that the metal and silicate in Watson were derived from distinct parent bodies that collided.

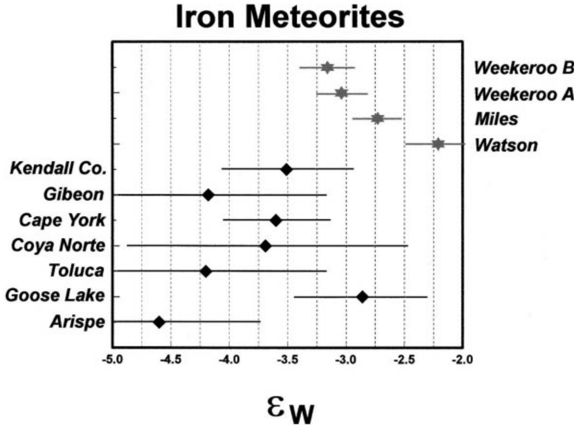

Fig. 3. Histogram of ε_W values for iron meteorites (data from [1] and this work). Note that, with the exception of Goose Lake which includes a large error, the IIE irons have the least negative ε_W values.

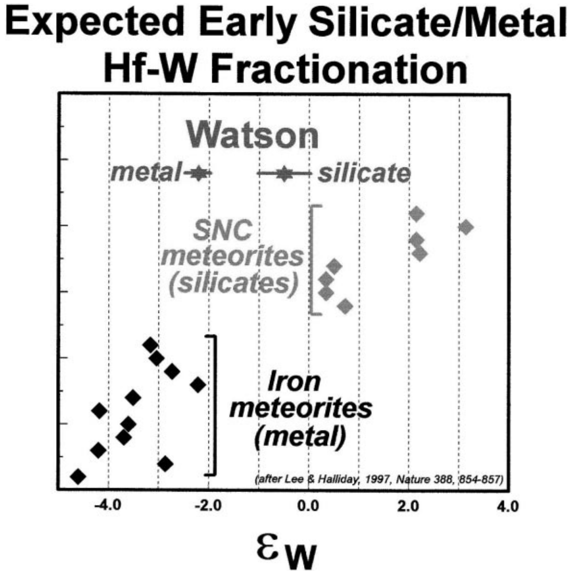

Fig. 4. Histogram of ε_W values for metals in iron meteorites, SNC meteorites (example of silicates which may have been in equilibrium with early FeNi metal fractionation), and the FeNi metal and silicate from the Watson IIE iron (data from [1,10], and this work).

7.2. *Petrogenesis of silicate inclusions*

A closer look at the Rb–Sr systematics versus the Sm–Nd systematics for these silicate inclusions yields some additional observations. First, there appears to be a relationship between the degree of LREE depletion in these inclusions and their Sm–Nd model ages (Fig. 5A). Specifically, as the T_{CHUR} ages of the IIE silicate inclusions become older, there is a corresponding increase in the ^{147}Sm/^{144}Nd (i.e., increasing LREE depletion) of the samples. However, such a corresponding relationship is not apparent for ^{87}Rb/^{86}Sr in these inclusions (Figs. 5B and 6), suggesting either that the Rb–Sr systematics are decoupled from the Sm–Nd systematics, or that the ^{87}Rb/^{86}Sr ratios have been altered, possibly during later impact metamorphism of the parent body.

7.2.1. *Weekeroo Station*

We have analyzed two separate inclusions from the Weekeroo Station IIE iron for Rb–Sr and Sm–Nd. The two inclusions indicate a complex petrogenetic history. Neither of the inclusions lies along the two lines defined by previous Rb–Sr isotopic data (Fig. 1A,B). If the ages of these two inclusions were considered to be similar to the previous analyses (e.g., [3,4]), then they would represent reservoirs with vastly different initial ^{87}Sr/^{86}Sr. In fact, silicate inclusion A, if 4.3–4.4 Ga old, would project back to an initial ^{87}Sr/^{86}Sr of 0.708, and inclusion B would project back to an initial ^{87}Sr/^{86}Sr which would approximate that of BABI (basalt achondrite best initial; 0.69899 ± 5; [48]) at this time.

Sm–Nd isotopic systematics of the Weekeroo Station silicate inclusions point to a complex and long-lived scenario. The simplest two-stage model for Weekeroo Station silicate inclusions A and B involves evolution in an H-chondrite parent body followed by LILE (i.e., Rb and Nd) enrichment at 700 Ma (Fig. 2). Such an enrichment event is also suggested by reconnaissance

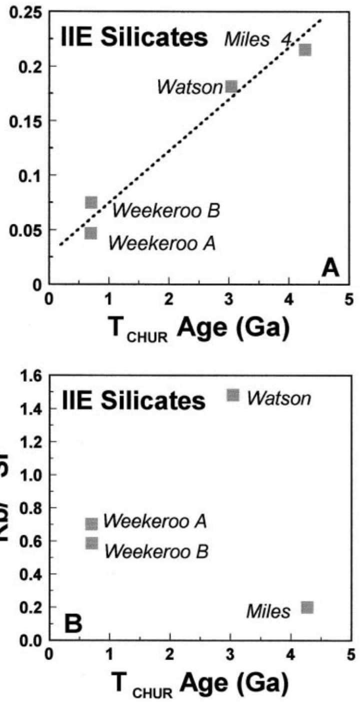

Fig. 5. Plots of parent–daughter ratios versus model T_{CHUR} ages for the four IIE silicate inclusions analyzed in this study. (A) ^{147}Sm/^{144}Nd versus T_{CHUR} ages showing a positive correlation. (B) ^{87}Rb/^{86}Sr versus T_{CHUR} ages showing no correlation.

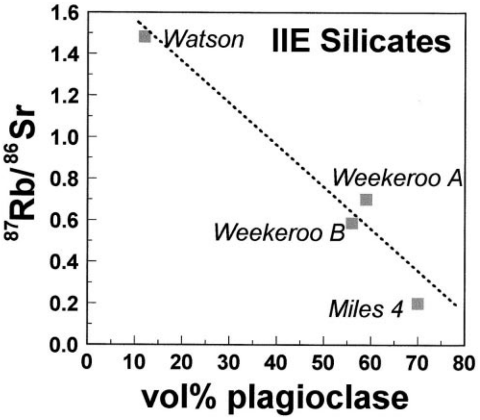

Fig. 6. ^{87}Rb/^{86}Sr versus the proportion of plagioclase (vol%) in the four IIE silicates studied.

Re–Os isotopic work [37]. Although other IIE irons yielded Re–Os model ages of 4.6 Ga, Weekeroo Station was the sole iron meteorite which gave an unrealistic Re–Os model age of 7.1 Ga. Such an impossibly old age could be due to enrichment in radiogenic Os or a decrease in the Re/Os ratio at some later time, possibly due to melting and loss of a sulfide and/or metal phase. This enrichment depletion event was likely due to impact on the parent body [29]. The effect of such an impact is seen most clearly by the occurrence of glass and cryptocrystalline silicates in the inclusions, probably caused by remelting and subsequent quenching of the inclusions (e.g., [6,9,29,38]). The impact also caused dynamic mixing of phases, as evidenced by the disequilibrium between augite and glass and the preferential alignment of large pyroxene grains [29]. Ruzicka et al. [29] discuss the possibility that igneous differentiation was impact-induced and conclude that the evidence for this is equivocal. The Sm–Nd isotopic systematics support the possibility of impact-induced differentiation by showing that LILE enrichment for Weekeroo Station inclusions A and B could have occurred as recently as 700 Ma ago, most likely during an impact melting episode.

7.2.2. Watson

Among the samples studied, the ^{147}Sm/^{144}Nd (0.1815) and present-day $\varepsilon_{\mathrm{Nd}}$ (−5.9) for Watson is the closest to that of chondrites (0.1967 and 0, respectively). Sm–Nd isotopic studies of the silicate in Watson indicate a LILE enrichment event, although of a lesser magnitude than that for Weekeroo Station, and much earlier in parent body evolution (at approximately 3040 Ma, Fig. 2). The ^{4}He–U–Th gas retention age for a split of the same large silicate inclusion from Watson (3.1 ± 0.3 Ga; [12]) is indistinguishable from the Sm–Nd model age of the silicate we analyzed (3.04 Ga). Olsen et al. [12] also analyzed K–Ar in their silicate, which yielded a slightly older age of 3.5 ± 0.2 Ga. Independently, all three of these isotopic systems indicate a late-fractionation event which occurred between 3 and 3.5 Ga ago.

The two Weekeroo Station samples are greatly LREE-enriched (^{147}Sm/^{144}Nd = 0.04646–0.07481; present-day $\varepsilon_{\mathrm{Nd}} = -11.0$ to -13.4) relative to chondrites. The silicate in Miles is also fractionated relative to chondrites, but in an opposite sense to both Watson and Weekeroo Station (^{147}Sm/^{144}Nd = 0.2154; $\varepsilon_{\mathrm{Nd}} = +10.3$). The recent work of Ebihara et al. [39] on Miles supports this conclusion. Based upon comparisons of the REE patterns and abundances in six gabbroic and three cryptocrystalline inclusions in Miles with those from other IIEs, they concluded that the parental liquid for Watson was the least differentiated and that for Weekeroo Station was the most evolved, with Miles having an intermediate composition.

Rb–Sr isotopes in the Watson silicate inclusion are opposite of what one might expect from the Sm–Nd isotopic studies. Miles, being the most LREE-depleted sample, also has the lowest Rb/Sr ratio, as would be expected. However, the two Weekeroo Station inclusions, being the most LREE-enriched, should also have the highest Rb/Sr. Clearly, their Rb/Sr ratios are higher than Miles, but Watson has a much higher Rb/Sr by a factor of two or more.

7.2.3. Miles

In contrast to that in Watson, the silicate in Miles points to an even earlier LILE depletion event in parent body evolution at 4270 Ma (Fig. 2). This rather imprecise Sm–Nd CHUR model age is, however, similar to the ^{39}Ar–^{40}Ar age of

4408 ± 11 Ma determined by Garrison and Bogard [15]. Although the Hf–W isotopic studies of Watson showed that the silicate inclusion was not in equilibrium with the enclosing metal, this may not be true for Miles. Similar siderophile-element patterns between the silicate inclusions and the host FeNi metal, as well as a relationship between the abundance of metal and the siderophile-element abundances in the silicate inclusions, led Ebihara et al. [39] to suggest that 'the two (Miles and Watson) appear to be genetically related'. Thus, we consider it likely that a major LILE depletion event occurred on the Miles IIE parent body between 4.3 and 4.4 Ga. This event appears to have either rehomogenized the silicate and the host FeNi metal, or caused the original separation of these phases. This fractionation event could have been caused by impact but, just as plausibly, could have been the result of igneous fractionation.

7.3. *Rb–Sr isochrons: true ages or mixing of impact components?*

Regardless of the complicated scenario one prefers for the origin of IIE iron meteorite silicate inclusions, two contrasting facts must be explained: (1) the Rb–Sr systematics from many other studies suggest a consistently very old age (4.3–4.5 Ga), whereas (2) the Sm–Nd systematics, with the exception of Miles-4, indicate progressively younger ages (Fig. 5A). The answer may lie in the impact processing which undoubtedly affected these meteorites. Shock-recovery experiments of basaltic materials have shown that the first phase to melt and form a glass is plagioclase feldspar, followed by pyroxene, and then olivine [40,41]. This feldspar-enriched melt would likely separate and coalesce with other feldspar-enriched droplets. Thus, progressive impact events can enrich a sample in the feldspar component, as has been shown for igneous-textured clasts in the Julesberg ordinary chondrite [42]. Such progressive enrichment in a feldspathic melt component would have important implications for the Rb–Sr and Sm–Nd systematics for silicate inclusions derived in a like manner.

Plagioclase contains vanishingly small abundances of Rb and will not generate much radiogenic Sr over an extended time period. Thus, resetting of the Sr isotopic systematics in the plagioclase at various times in the history of a periodically impacted sample would not change its placement on a Rb–Sr diagram, like that in Fig. 3. The 'ages' generated from a sample that has been multiply impacted and has remelted plagioclase several times in its history would not be changed, but would likely retain its original crystallization age (i.e., the plagioclase plots very near or practically on the *y*-axis and cannot be easily moved from this axis). Thus, the placement of samples on Fig. 1 simply could reflect the mineralogy of the samples, and the derivative line could be a mixing line between mafic minerals and remelted plagioclase and glass.

The Weekeroo Station samples exhibit petrographic evidence which is consistent with this model of preferential feldspar melting. Coarse orthopyroxene exsolution lamellae in Weekeroo Station B pyroxenes are suggestive of slow-cooling, possibly as a result of deep burial in the parent body. In contrast, the cryptocrystalline feldspar+silica glass in Weekeroo Station A is indicative of rapid, likely near-surface, cooling. The preservation of this coarse exsolution lamellae in Weekeroo Station B suggests that the pyroxenes were not remelted during the event (impact?) that created the glass in Weekeroo Station A.

Kodaikanal is the only IIE iron to be analyzed that yielded a significantly younger Rb–Sr age (e.g., 3743 ± 44 Ma) [3,4]. If our scenario of preferential plagioclase melting is correct, then this sample would suggest that a shock event must have been intense enough that other phases were also melted, possibly totally resetting all of the phases. Just such an intense event is recorded in the FeNi metal of Kodaikanal. In fact, Buchwald [43] stated that 'few iron meteorites display such distortions (of silicate–metal, troilite–metal, and schreibersite–metal interfaces) as those present in Kodaikanal'. He goes on to state that these distortions and deformation features 'require shock with attenuated peak temperatures for their formation' [43].

In mafic rocks, olivine and pyroxenes typically exhibit the highest Sm/Nd of any of the major phases and plagioclase the lowest Sm/Nd. With the exception of augite, plagioclase also exhibits the highest abundance of Nd among the major phases. However, the Sm/Nd of plagioclase is not vanishingly small, and radiogenic Nd would be expected to build up, given a certain period of time. Furthermore, augite is a significant, high-Nd, high-Sm/Nd component, which also can melt with plagioclase during an intense impact. Thus, determining the Sm–Nd distribution during impact melting is exceedingly complicated, and it is much more likely the Sm–Nd systematics would be at least partially reset with each successive event. Thus, whereas the Rb–Sr systematics would not yield any information other than the original igneous fractionation event, the Sm–Nd systematics could reflect the impact history of the sample. Supporting evidence for this is found in the Sm–Nd model (T_{CHUR}) ages, which increase with increasing LREE depletion of the sample (Table 2). Such a relationship might be expected, if successive impact events create more and more feldspathic glass, with increasingly enriched LREE.

8. Models for IIE silicate formation

The lack of equilibration between silicate and FeNi metal in the Watson IIE iron meteorite strongly suggests that the enclosed silicate formed much later than the FeNi host. Thus, it appears that the parent body for the IIE FeNi metal fractionated silicate from metal early (to achieve the W isotopic fractionation seen in the FeNi metal), but that this silicate is no longer present. The silicate that is enclosed within the FeNi metal was introduced later, in some cases up to 1 billion yr later.

The silicates enclosed by the FeNi metal have undergone variably complex histories. If these silicate inclusions were all derived from H-group ordinary chondrites, as is currently thought, then these silicates suggest fractionation events which span over 3 billion yr. In some cases, as in Miles, the silicates represent the LREE-depleted crystallization products of a melt generated within a few hundred million years (4.3–4.4 Ga ago) of formation of the FeNi metal. Silicate inclusions in Watson probably are imperfectly separated residues of partial melting of a chondritic precursor 3–3.5 Ga ago. Silicate inclusions in Weekeroo Station point to a recent event, only 700 Ma ago, which generated very evolved melts. All of these fractionation events could have been initiated by near-surface processes, presumably impacts and/or collisions and consequent mixing, as is suggested through textural and mineral–chemical studies [6,7,12,29,30,44].

Ruzicka et al. [29] presented two models for the formation of IIE meteorites. Both models include collision and mixing between a metal impactor and silicate target, but the difference is whether the target was represented by previously fractionated or primitive H-chondrite silicate. Due to the large Rb/Sr fractionations seen for IIE silicates, and especially in the Watson and Weekeroo Station silicates, we consider it unlikely that the precursor was undifferentiated. Instead, it is much more feasible to fractionate an H-chondrite precursor in several consecutive stages.

The metals in the IIE parent body were fractionated from a silicate mass early in its history, however, this silicate is either not present in the IIE iron meteorites (and by conjecture not in the parent body) or has been significantly reprocessed. More importantly, the isotopic systematics of the IIE irons suggest a long history of processing of the silicate portion of the parent body, possibly continuing up until 700 million yr ago.

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