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Space weathering processes on airless bodies: Fe isotope fractionation in the lunar regolith

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Abstract

Nanophase Fe metal grains (np-Fe⁰) are a product of space weathering, formed by processes related to meteorite impacts, and solar-wind sputtering on airless planetary bodies, such as the Moon. Iron isotopes of lunar soils are fractionated during these processes, and the np-Fe⁰ in the finest (< 10 μm), mature, size fractions of the soil become enriched in heavier isotopes by ~0.3‰ in ⁵⁶Fe/⁵⁴Fe in comparison to the bulk rocks (0.03 ± 0.05‰), from which the soil was formed. A positive correlation of δ⁵⁶Fe values with the soil maturity index, I_S/FeO, suggests that the high δ⁵⁶Fe values reflect production of nanophase Fe metal that is produced by space weathering that occurs on airless planetary bodies. Furthermore, the enrichment of δ⁵⁶Fe in the smallest size fraction of lunar soils supports a model for creation of np-Fe⁰ through vapor deposition induced by micrometeorites, as well as that by solar-wind sputtering. © 2003 Elsevier B.V. All rights reserved.

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1. Introduction

Understanding the effects of space weathering on the surfaces of airless planetary bodies such as the Moon, Vesta, Phobos, etc., is important in interpreting remote-sensing data. For example, alteration of the lunar regolith by meteorite and micrometeorite impacts, solar-wind and flare particles, and cosmic rays masks and hinders effective

use of remote sensing by spectral reflectance [1,2]. The optical properties of lunar soils are altered during space weathering in that the soils, in comparison with freshly ground lunar rocks, become darker, absorption bands are weaker, and develop a ‘reddened’ spectrum [1]. Specifically, the presence of major amounts of nanophase Fe metal grains (np-Fe⁰) in the < 10 μm portion of the lunar soil adversely affects deciphering of the optical properties (e.g. chemistry, mineralogy) of lunar soils [1]. In addition, the effects of space weathering have been evoked to explain the observed spectral mismatch between the S-type asteroids and ordinary chondrites [3–5].

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Studies of lunar soils have documented that np-Fe^o is present in the glassy portions of the soils and mainly in aggregates of minerals, rocks, and glasses called agglutinates. It is the glass that binds the soil fragments together that contains appreciable np-Fe^o. Rims of lunar soil particles also contain substantial amounts of np-Fe^o, as has been demonstrated by Keller et al. [6]. Although it has been generally assumed that np-Fe^o is the result of reduction of FeO in impact melts by solar-wind-implanted hydrogen [7], several difficulties with this scenario have never been resolved, such as the lack of evidence for H₂O in lunar soils [8]. In addition, laboratory simulations of space weathering using pulsed laser beams have shown that the presence of H is not a requirement for the formation of np-Fe^o [9,10]. Another source and mechanism of formation for np-Fe^o, although predicted in Apollo days [11], has been emphasized the last several years [12]. Upon impact melting of lunar soil, some silicate melt is also vaporized, selectively releasing volatile elements into the vapor phase. In addition to the volatile elements S, Na, and K, FeO and SiO₂ are also vaporized, where FeO can undergo further dissociation to elemental Fe [4]. Volatilized Fe and SiO₂ and other oxides condense on the surfaces of virtually all soil particles and form thin layers that contain np-Fe^o [6]. Recent transmission electron microscope (TEM) studies of lunar regolith samples support a model for vapor-phase deposition of np-Fe^o. These TEM studies were performed on <20 μm size fractions of lunar soils and demonstrated that np-Fe^o occurs within the upper 100 nm of the surface of many regolith grains [6,12]. Moreover, np-Fe^o occurs on both ferrous-poor and ferrous-rich mineral and silicate-glass grains in near equal proportions [13]. These vapor-deposited coatings are seen as an additional, significant and important source of np-Fe^o, in addition to that possibly produced by the solar-wind hydrogen autoreduction process [2,14].

Lunar regolith samples display mass-dependent, heavy-isotope enrichments of O, Si, S, K, Cd, Ca, and Mg, relative to rocks collected from the lunar surface [15–21]. The processes that cause these unidirectional, kinetic-isotope fractionations in-

clude ion sputtering, vaporization by meteorite impact, and redeposition of sputtered and volatilized matter following gravitational fractionation, and are responsible for modifying the lunar regolith [22,23]. These isotopic studies were conducted on bulk lunar regolith samples, hence, it is difficult to infer if the preferential destruction or production of a specific phase was the cause for the mass-dependent isotopic variations. Some studies used partial dissolution techniques to identify the phase or phases that recorded these isotopic variations, and found that the heaviest isotope enrichments occur in the initial extraction [15,20,21]. However, it has been noted that the large isotopic effects observed for O and Si might be artifacts of analytical procedures [21].

In this study, we report on iron isotope variations of lunar rocks and selected size fractions of several mare soils to determine if space weathering produces a distinct isotopic signature in lunar soils with different maturities (i.e. different exposure times at the lunar surface). It will be shown that surface processes (space weathering), indeed, do modify Fe isotopes in lunar soils. Specifically, the likely process responsible for Fe isotopic fractionation is vaporization of regolith matter during micrometeorite impacts, although Fe isotopes may also be fractionated by solar-wind sputtering.

2. Materials and methods

The lunar regolith samples analyzed in this study have been previously characterized (mineralogy, modal abundance, spectral reflectance, and magnetic properties) by the Lunar Soil Characterization Consortium [2]. Additionally, the relative amounts of np-Fe^o were determined for each soil fraction based on their I_S/FeO values, where I_S is the ferromagnetic resonance (FMR) intensity signal of the near single-domain (i.e. np-Fe^o), elemental Fe particles (3–40 nm) [24]. The FMR intensity is normalized to the total FeO content of the soil fraction (I_S/FeO), to give an approximate value for the fraction of the total iron in a sample that is in the np-Fe^o state. This factor is a function of exposure of the soil at the lunar surface and is used as the ‘maturity index’ for all lunar

soils, where values of 30 and 60 delineate ‘immature’ from ‘submature’, and ‘submature’ from ‘mature’, respectively [24]. A portion of the np-Fe^o in the soil fractions is present in the agglutinitic glass. However, an even larger proportion of the np-Fe^o signal, particularly in the finest size fractions, is attributed to the surface-correlated, vapor-deposited np-Fe^o, which increases as the surface area to volume ratio increases [2].

The Lunar Soil Characterization Consortium samples were sieved into sized fractions (<10, 10–20, and 20–45 μm) [2], and six of these mare soils were analyzed for their Fe isotope composition. Furthermore, the smallest size fraction (<10 μm) of soil 79221 was partially leached in 1 M HCl, and the leachate (63% of the total Fe) and the residue were analyzed for their Fe isotope composition. All Fe isotope analyses were performed using the University of Wisconsin Micro-mass *IsoProbe*. The analytical methods for Fe isotope analyses followed the methods reported by Skulan et al. [25] and Beard et al. [26]. Iron isotope compositions from a variety of terrestrial igneous volcanic and plutonic rocks (Terr Ig rocks) are homogeneous within ±0.05‰ (1σ) and form a natural baseline to which Fe isotope variations may be compared [26]. Based on replicate analyses of samples processed through the entire analytical procedure two or more times, the average *external* reproducibility (1σ, S.D.) for the ⁵⁶Fe/⁵⁴Fe ratio is ±0.05‰. Analyses are reported using delta notation, where δ⁵⁶Fe = (⁵⁶Fe/⁵⁴Fe)_{Sample} / (⁵⁶Fe/⁵⁴Fe)_{Terr Ig rocks} - 1) 10³ and δ⁵⁷Fe = (⁵⁷Fe/⁵⁴Fe)_{Sample} / (⁵⁷Fe/⁵⁴Fe)_{Terr Ig rocks} - 1) 10³. On this scale, the measured Fe isotope composition of the IRMM-014 Fe isotope standard was δ⁵⁶Fe = -0.09 ± 0.05‰ and δ⁵⁷Fe = -0.11 ± 0.07‰.

3. Fe isotope compositions of lunar soils and rocks

The results of Fe isotope analyses of different bulk soils are plotted against the I_S/FeO value for the size fractions of each soil (Fig. 1, Table 1). There is a positive correlation between I_S/FeO (which is proportional to the amount of np-Fe^o) and δ⁵⁶Fe values. In general, values of I_S/FeO

increase with decreasing grain size and exposure time on the lunar surface [2,24]. Soils 71061 and 12030 (immature) show no correlation with isotopic composition. The <10 μm size fraction of the mature soil 79221 has the highest I_S/FeO value, and this sample also has the highest δ⁵⁶Fe value (+0.34 ± 0.05‰). On a δ⁵⁷Fe–δ⁵⁶Fe plot (Fig. 2), all analyses plot along a line with a slope of 2/3, indicating that the measured fractionations are associated with mass-dependent processes.

In addition to the analyzed lunar regolith samples, we have reanalyzed the five Apollo 12 mare basalt samples reported in [27], as well as three additional mare basalts (two from Apollo 12 and one from Apollo 17), and one impact-melt breccia from the Apollo 16 highlands (Fig. 3). The δ⁵⁶Fe value of the Apollo 17 high-Ti basalt is +0.13 ± 0.02‰, which is similar to the Fe isotope composition of lunar samples reported by Poitrasson et al. [28], i.e. +0.07 ± 0.04‰ (data corrected through a common value for the IRMM-014 Fe isotope standard). In contrast, our analyses of Apollo 12 rocks (low-Ti mare basalts) and the Apollo 16 impact rock yield lower δ⁵⁶Fe values of +0.02 ± 0.04‰ and -0.04 ± 0.02‰, respectively (Table 1).

The Fe isotope compositions of eight lunar mare basalts and one highland rock have δ⁵⁶Fe values that range from -0.04 to +0.13‰, with a mean of 0.03 ± 0.05‰. This is a range similar to the mean of terrestrial igneous rocks (0.00 ± 0.05‰, 1σ) [25,26]. In contrast, Poitrasson et al. [28,29] reported that δ⁵⁶Fe of the Earth and Moon are distinct, where the δ⁵⁶Fe of the Earth is ~0.07‰ lower than that of the Moon. Immature soils 71061 and 12030, which have size fractions with I_S/FeO values <33, also have Fe isotope compositions that overlap those of the average for lunar rocks, indicating that these soils have not had sufficient time to develop isotopically distinctive np-Fe^o compositions. In contrast, the mature lunar soils 79221 and 15041, specifically the <10 μm size fractions, have I_S/FeO values of 169 and 161, respectively, and have δ⁵⁶Fe values that are higher than those of lunar basalts by a factor of two to three. This positive correlation between I_S/FeO and δ⁵⁶Fe values is taken as strong evidence that the lunar regolith phase that

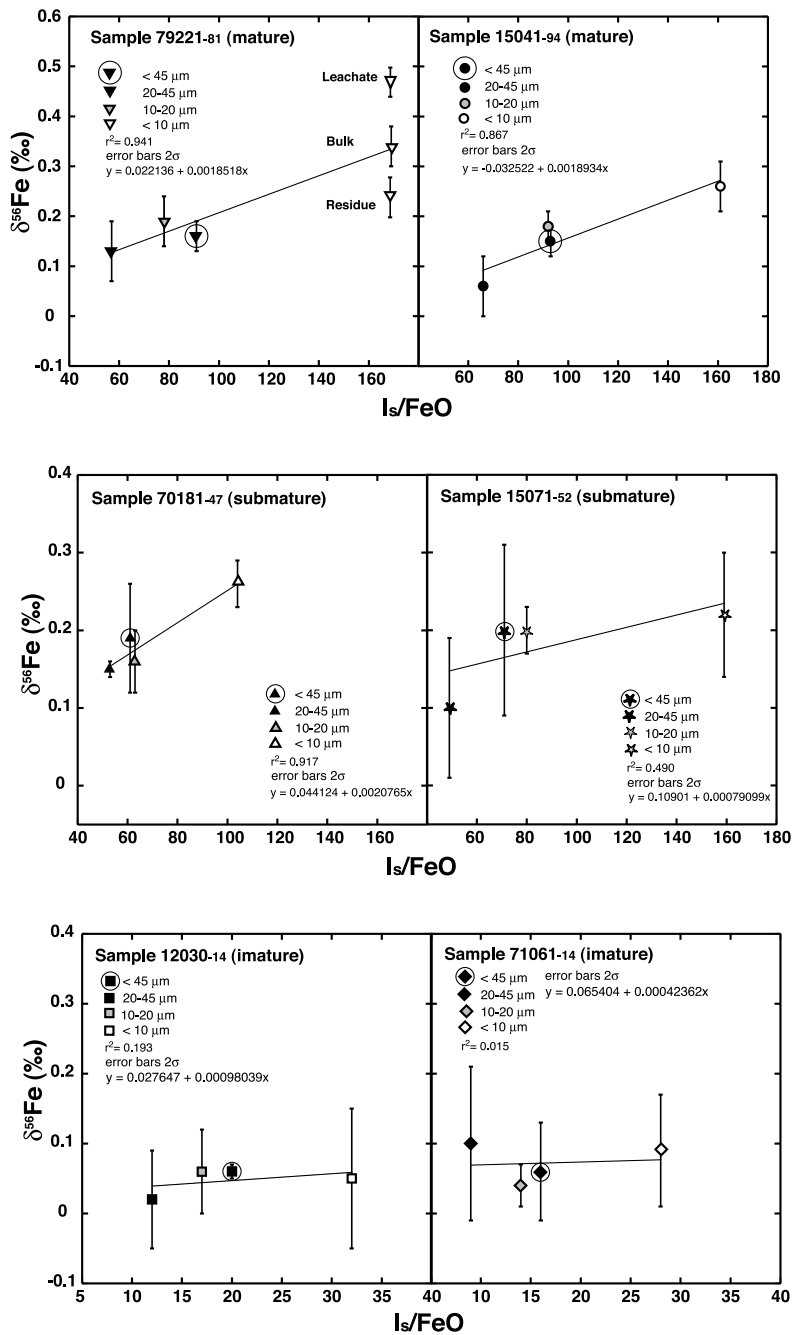


Fig. 1. Plot of I_S/FeO vs. $\delta^{56}\text{Fe}$ for lunar soils. Numbers after samples indicate I_S/FeO of $<250 \mu\text{m}$ size fraction. All analyses are for bulk samples of different size fractions. The size fraction $<45 \mu\text{m}$ is the original bulk sample before sieving. In addition, the $<10 \mu\text{m}$ size fraction of 79221 has been leached and the $\delta^{56}\text{Fe}$ values of the leachate and the residue have been plotted with the same I_S/FeO as the bulk sample, although I_S/FeO may be different for the leachate and the residue. Error bars are 2σ (2 standard error) from two or more mass spectrometry analyses.

has high $\delta^{56}\text{Fe}$ values is np-Fe^o. In addition, the smallest size fractions of the lunar soil have the highest surface to volume ratio, implying that most of the np-Fe^o metal is located on surface of the soil particles. The 1 M HCl leaching experiment performed on the <10 μm size fraction of the mature soil sample 79221 is consistent with this hypothesis. Because HCl preferentially attacks Fe metal, the leachate $\delta^{56}\text{Fe}$ value of +0.47‰ (63 mol% of the total Fe in this sample) is interpreted to contain the greatest proportion of np-Fe^o. Using the residue ($\delta^{56}\text{Fe} = +0.21\text{‰}$), and assuming a linear relationship between the Fe isotope compositions of the bulk and the residue of soil 79221 (<10 μm), and the $\delta^{56}\text{Fe}$ of the leachate, the projected Fe isotope composition of the pure np-Fe^o component would be +0.7‰.

Meteorite influx that contains extralunar iron phases could potentially alter iron isotope composition of lunar soils. Lunar soils from the mare regions contain on the average ~ 1 wt% metallic Fe [30], and it is estimated that $\sim 30\%$ of the large-sized native Fe in lunar soils is of meteoritic origin [30]. The bulk of meteoritic material hitting the Moon and the Earth is chondritic and most closely resembles CM chondrite bulk compositions (e.g. [31,32]). Bulk Fe isotope analysis of a sample from a CM2 chondrite (Murchison) yielded $\delta^{56}\text{Fe} = -0.09\text{‰}$ [33] (data corrected through a common value for the IRMM-014 Fe isotope standard). Iron meteorites, which only

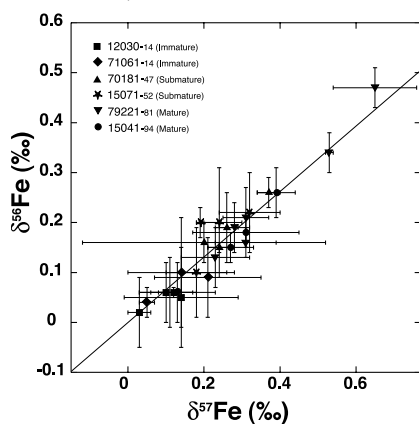


Fig. 2. Plot of $\delta^{57}\text{Fe}$ vs. $\delta^{56}\text{Fe}$ for the lunar mare soils. Small numbers after the samples indicate I_s/FeO for the <250 μm size fraction. Error bars show 2σ (2 standard error) from two or more mass spectrometry analyses.

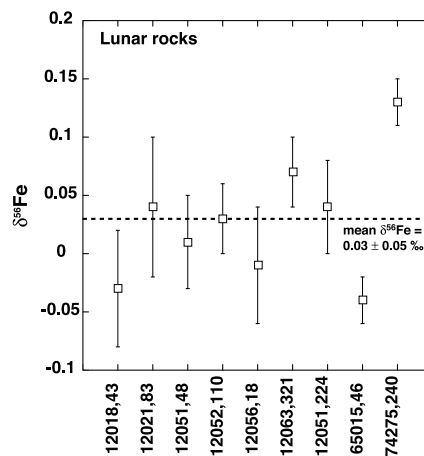


Fig. 3. Fe isotope compositions of various lunar rocks. Samples 12018,43, 12021,83, 12051,48, 12052,110, 12056,18, 12063,321, and 12051,224 are low-Ti mare basalts. Sample 65015,46 is an impact-melt rock and sample 74275,240 is a high-Ti mare basalt. Error bars indicate 2σ (2 standard error).

contribute $\sim 5\%$ of meteorite falls [34], define a relatively small range in Fe isotope composition, where $\delta^{56}\text{Fe}$ values vary from -0.07 to $+0.06\text{‰}$ for the seven iron meteorites that have been analyzed to date [33]. From this limited range, we conclude that it is unlikely that the trend we observe in our analyses could be produced through a net Fe addition by a meteoritic contribution.

4. Mechanisms of Fe isotope fractionation

The surfaces of lunar soil grains are modified by vapor deposition from impact melts during micrometeorite shock events and by irradiation with solar-wind particles. The relative contributions of the two mechanisms have been contentious [12,35–38]. Although the production of amorphous rims on lunar soil grains has been assumed to be a result of solar-wind irradiation [35,39], recent investigations using TEM have revealed that rims of lunar soil grains are not homogeneous and contain a variety of different morphologies and chemical compositions [6]. It has been shown that vapor deposition on individual soil grains can also be a major factor in producing the ubiquitous patinas and rims that contain sili-

Table 1
Fe isotope composition of lunar regolith samples

Sample	Size fraction	I_S/FeO	$\delta^{56}\text{Fe}$	2σ	$\delta^{57}\text{Fe}$	2σ	n
Lunar soils							
12030,156	< 45 μm	20	0.06	0.01	0.12	0.06	2
12030,157	20–45 μm	12	0.02	0.07	0.03	0.03	2
12030,158	10–20 μm	17	0.06	0.06	0.10	0.07	4
12030,159	< 10 μm	32	0.05	0.10	0.14	0.16	2
71061,150	< 45 μm	16	0.06	0.07	0.11	0.03	2
71061,158	20–45 μm	9	0.10	0.11	0.14	0.14	3
71061,163	10–20 μm	14	0.04	0.03	0.05	0.02	3
71061,168	< 10 μm	28	0.09	0.08	0.21	0.14	2
70181,147	< 45 μm	61	0.19	0.07	0.26	0.03	2
70181,155	20–45 μm	53	0.15	0.01	0.24	0.03	2
70181,160	10–20 μm	63	0.16	0.04	0.20	0.22	2
70181,165	< 10 μm	104	0.26	0.03	0.37	0.03	2
15071,167	< 45 μm	71	0.20	0.11	0.24	0.06	2
15071,168	20–45 μm	49	0.10	0.09	0.18	0.08	3
15071,169	10–20 μm	80	0.20	0.03	0.19	0.01	2
15071,170	< 10 μm	159	0.22	0.08	0.32	0.08	3
79221,118	< 45 μm	91	0.16	0.04	0.36	0.00	2
			0.15	0.07	0.26	0.14	2
79221,126	20–45 μm	57	0.13	0.11	0.25	0.11	2
			0.12	0.10	0.21	0.17	2
79221,131	10–20 μm	78	0.19	0.02	0.27	0.02	3
			0.20	0.11	0.29	0.08	3
79221,136	< 10 μm	169	0.32	0.05	0.53	0.00	3
			0.35	0.08	0.54	0.01	2
79221,136	leachate	169	0.47	0.04	0.65	0.11	2
79221,136	residue	169	0.21	0.06	0.31	0.06	2
15041,221	< 45 μm	93	0.14	0.06	0.24	0.09	3
			0.15	0.01	0.30	0.06	2
15041,222	20–45 μm	66	0.09	0.02	0.20	0.06	3
			0.02	0.11	0.05	0.16	3
15041,223	10–20 μm	92	0.16	0.06	0.24	0.22	2
			0.20	0.03	0.38	0.17	2
15041,224	< 10 μm	161	0.26	0.01	0.40	0.00	2
			0.30	0.11	0.43	0.06	2
			0.22	0.11	0.34	0.13	2
Lunar rocks							
12018,43	low-Ti mare basalt		–0.07	0.07	–0.05	0.06	2
			–0.03	0.04	0.06	0.13	2
			–0.01	0.10	0.05	0.13	3
12021,83	low-Ti mare basalt		0.01	0.08	0.05	0.04	1
			0.05	0.08	0.05	0.06	3
12051,48	low-Ti mare basalt		0.00	0.09	0.03	0.09	3
			0.02	0.01	0.02	0.08	2
12052,110	low-Ti mare basalt		0.00	0.03	0.04	0.01	2
			0.05	0.00	0.02	0.08	2
12056,18	low-Ti mare basalt		–0.04	0.10	–0.10	0.07	2
			–0.05	0.10	0.06	0.05	1
			0.04	0.01	0.03	0.01	2
12063,321	low-Ti mare basalt		0.07	0.03	0.12	0.07	3
12051,224	low-Ti mare basalt		0.04	0.04	0.07	0.03	2
65015,46	impact-melt rock		–0.06	0.00	–0.10	0.03	2
			–0.02	0.01	–0.04	0.01	2
74275,240	high-Ti mare basalt		0.13	0.02	0.16	0.02	3

The size fraction < 45 μm is the original bulk sample before sieving. Each row represents a separate dissolution and chemical processing through the ion-exchange columns. The isotopic results are averages of n duplicate isotope analyses. Errors are 2σ of two or more mass spectrometry analyses, except for samples 12056,18 and 12021,83 ($n=1$), where the error is 2σ from the internal counting statistics.

ca-rich, probably amorphous, glass with embedded np-Fe⁰ [6,12]. Various models and experiments predict that chemical and isotopic fractionation should occur during these processes (e.g. [4,22,23,40–42]).

Our Fe isotope data clearly demonstrate that there are significant Fe isotope variations in the lunar regolith, and that this isotopic variability is related to surface-correlated processes. The occurrence of nanometer-sized, spherical particles of Fe metal in the rims of soil grains, particularly on iron-free mineral grains (e.g. plagioclase), requires that these np-Fe⁰ spherules were added to the grain surface by vapor deposition. Therefore, it is envisioned that Fe may be fractionated by a variety of processes that occur during micrometeorite impacts, such as vaporization of FeO, dissociation to elemental Fe, or condensation, producing heavier $\delta^{56}\text{Fe}$ values in the smallest size fraction of mature soils. Mass balance requires a reservoir of isotopically light Fe, which is most likely to have been lost to space. In contrast, formation of np-Fe⁰ by auto reduction of impact melts with H is not likely to produce Fe isotope variation in a bulk analysis, unless the Fe is vaporized.

Another process by which Fe isotopes may be fractionated is through sputtering by solar-wind irradiation. There is ample evidence that solar-wind irradiation produces amorphous rims on lunar soil grains [6,35,39], although the extent of rim formation in part seems to depend on the nature of the grains (silicates versus oxides) [43]. Irradiation of mafic composition lunar grains (e.g. pyroxene, olivine) preferentially removes Mg and Ca relative to other ions during irradiation, leaving behind an amorphous rim enriched in O [44]. Experimental sputtering of Ca minerals demonstrated that the lighter isotopes of Ca are preferentially removed from the irradiated surface [45]. Additional isotopic fractionation may occur during gravitational settling and by angular dependence of the fractionation [45]. In contrast, Ca isotope fractionation measured from lunar soils (extracted by leaching) yielded only up to 1‰ amu⁻¹, which is a much smaller fractionation than has been observed during sputtering experiments [20]. It has been suggested that experimen-

tal problems during Ca extraction (dilution and preferential removal) [21] may have caused the small Ca isotope fractionation measured in lunar soils. Mass balance requires that if all sputtered material is incorporated into nearby soils, there should be no net fractionation observable, and this same argument is applicable to fractionation during micrometeorite impacts. However, our Fe isotope data, as well as work on other isotope systems in lunar soils (e.g. [46]), suggest that some fractionation occurs due to the preferential loss of light isotopes to space. Although the contribution to vapor deposits comes from two competing processes, solar-wind sputtering and micrometeorite impacts, their relative amounts are not well constrained [6].

Previous isotope studies of lunar regolith samples have shown that mature soils are enriched in heavy isotopes of the metallic elements Cd, S, K, Si, Ca, and Mg [15,17–21] as compared to the isotopic composition of lunar igneous rocks. In general, the degree of isotopic fractionation is greatest (6–10‰ amu⁻¹, respectively) for elements that have low vaporization temperatures (e.g. Cd, S, and K) [17–19], whereas for metals that have higher vaporization temperatures (e.g. Mg and Ca), the isotopic fractionations are much smaller (1.6 and 0.8‰ amu⁻¹) [20,21]. Silicon is the exception to the general trend of increasing isotopic fractionation with lower vaporization temperature; Si has a vaporization temperature that is intermediate to Mg and Ca, but Si shows the largest isotopic fractionation of any of the metals (12.6‰ amu⁻¹) [15]. Direct comparison of the relative extent of isotopic fractionation among different elements is difficult because the maximum fractionations quoted above for different elements were determined on different soil samples, and employed different leaching methods. That is, the mixing of determinations would make for internally inconsistent data comparison.

Since the relative volatility of Fe during evaporation is higher than that of Mg [47,48], it is expected that Fe isotope fractionation should be larger or at least similar to that of Mg. The maximum Mg isotope fractionation observed is 1.7‰ amu⁻¹ for a lunar soil that was subjected to H₂O leaching, which extracted < 1 mol% of the total

Mg in bulk soil 66081 ($I_S/\text{FeO} = 80$) [21]. We have inferred an 0.35‰ amu^{-1} Fe isotope fractionation based on a linear extrapolation of a bulk analysis and leaching experiment from sample 79221,136. The linear extrapolation we use probably results in a minimum estimate of the Fe isotope composition of the np-Fe° , because most leaching studies define a logarithmic function of isotopic composition versus the percent of element leached from the soil [15].

5. Conclusions

The enrichment of $\delta^{56}\text{Fe}$ in the smallest-size fraction of mature soils is a signature of space weathering processes that operate on the lunar surface. The generally accepted view that the majority of the np-Fe° forms as a result of impact-generated melting of solar-wind-saturated soils has recently been questioned [4,49]. It has been speculated that the majority of np-Fe° was formed by vapor deposition and subsequent reworking of the soil [49]. Fractionation of Fe isotopes was most likely the result of some Fe vaporization, thus supporting this new paradigm. Moreover, our results highlight the importance of space weathering processes in producing mass-dependent isotopic fractionation of Fe, which indicates that vaporization and condensation processes can produce Fe isotope variability in extraterrestrial samples.

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References

- [1] C.M. Pieters, L.A. Taylor, S.K. Noble, L.P. Keller, B.W. Hapke, R.V. Morris, C.C. Allen, D.S. McKay, S. Wentworth, Space weathering on airless bodies: resolving a mystery with lunar samples, *Meteorit. Planet. Sci.* 35 (2000) 1101–1107.
- [2] L.A. Taylor, C.M. Pieters, L.P. Keller, R.V. Morris, D.S. McKay, Lunar mare soils: Space weathering and the major effect of surface-correlated nanophase Fe, *J. Geophys. Res.* 106 (2001) 27985–27999.
- [3] C.R. Chapman, S-type asteroids, ordinary chondrites, and space weathering: the evidence from Galileo's fly-bys of Gaspra and Ida, *Meteorit. Planet. Sci.* 31 (1996) 699–725.
- [4] B.W. Hapke, Space weathering from Mercury to the asteroid belt, *J. Geophys. Res.* 106 (2001) 10039–10073.
- [5] B.E. Clark, B.W. Hapke, C. Pieters, D. Britt, Asteroid space weathering and regolith evolution, in: W.F. Bottke, A. Cellino, P. Paolicchi, R.P. Binzel (Eds.), *Asteroids III*, University of Arizona Press, 2002, pp. 585–599.
- [6] L.P. Keller, D.S. McKay, The nature and origin of rims on lunar soil grains, *Geochim. Cosmochim. Acta* 61 (1997) 2331–2341.
- [7] R.M. Housley, R.W. Grant, N.E. Paton, Origin and characteristics of excess Fe metal in lunar glass welded aggregates, *Proc. 4th Lunar Sci. Conf.*, 1973, pp. 2737–2749.
- [8] L.A. Taylor, G.R. Rossman, Q. Qi, Where has all the lunar water gone? *Lunar Planet. Sci. Conf. XXVI*, 1995, pp. 1399–1400.
- [9] M. Yamada, S. Sasaki, H. Nagahara, A. Fujiwara, S. Hasegawa, H. Yano, T. Hiroi, H. Ohashi, H. Otake, Simulation of space weathering of planet-forming materials nanosecond pulse laser irradiation and proton implantation on olivine and pyroxene samples, *Earth Planets Space* 51 (1999) 1255–1265.
- [10] S. Sasaki, K. Nakamura, Y. Hamabe, E. Kurahashi, T. Hiroi, Production of iron nanoparticles by laser irradiation in a simulation of lunar-like space weathering, *Nature* 410 (2001) 555–557.
- [11] B.W. Hapke, W.A. Cassidy, E. Wells, Effects of vapor-phase deposition processes on the optical, chemical, and magnetic properties of the lunar regolith, *Moon* 13 (1975) 339–353.
- [12] L.P. Keller, D.S. McKay, Discovery of vapor deposits in the lunar regolith, *Science* 261 (1993) 1305–1307.
- [13] L.A. Taylor, A. Patchen, D.-H. Taylor, J.G. Chambers, D.S. McKay, X-ray digital imaging and petrography of lunar mare soils data input for remote sensing calibrations, *Icarus* 124 (1996) 500–512.
- [14] L.A. Taylor, C.M. Pieters, L.P. Keller, R.V. Morris, D.S. McKay, A. Patchen, S. Wentworth, The effects of space weathering on Apollo 17 mare soils: petrographic and chemical characterization, *Meteorit. Planet. Sci.* 36 (2001) 285–299.
- [15] S. Epstein, H.P. Taylor, $\text{O}^{19}/\text{O}^{16}$, $\text{Si}^{30}/\text{Si}^{28}$, D/H, and $\text{C}^{13}/\text{C}^{12}$ ratios in lunar samples, *Proc. 2nd Lunar Sci. Conf.*, 1971, pp. 1421–1441.
- [16] R.N. Clayton, T.K. Mayeda, J.M. Hurd, Loss of oxygen, silicon, sulfur, and potassium from the lunar regolith, *Proc. 5th Lunar Sci. Conf.*, 1974, pp. 1801–1809.

- [17] H.G. Thode, C.E. Rees, Sulphur isotopes in grain size fractions of lunar soils, *Proc. 7th Lunar Sci. Conf.*, 1976, pp. 459–468.
- [18] M. Humayun, R.N. Clayton, Precise determination of the isotopic composition of potassium: application to terrestrial rocks and lunar soils, *Geochim. Cosmochim. Acta* 59 (1995) 2115–2130.
- [19] D.G. Sands, K.J.R. Rosman, J.R. Laeterde, A preliminary study of cadmium mass fractionation in lunar soils, *Earth Planet. Sci. Lett.* 186 (2001) 103–111.
- [20] W.A. Russell, D.A. Papanastassiou, T.A. Tombrello, S. Epstein, Ca isotope fractionation on the moon, *Proc. 8th Lunar Sci. Conf.*, 1977, pp. 3791–3805.
- [21] T.M. Esat, S.R. Taylor, Magnesium isotope fractionation in lunar soils, *Geochim. Cosmochim. Acta* 56 (1992) 1025–1031.
- [22] R.M. Housley, A model for chemical and isotopic fractionation in the lunar regolith by impact vaporization, *Proc. 10th Lunar Planet. Sci. Conf.*, 1979, pp. 1673–1683.
- [23] Z.E. Switkowski, P.K. Haff, T.A. Tombrello, D.S. Burnett, Mass fractionation of the Lunar surface by solar wind sputtering, *J. Geophys. Res.* 82 (1977) 3797–3804.
- [24] R.V. Morris, Surface exposure indices of lunar soils: a comparative FMR study, *Proc. 7th Lunar Planet. Sci. Conf.*, 1976, pp. 315–335.
- [25] J.L. Skulan, B.L. Beard, C.M. Johnson, Kinetic and equilibrium Fe isotope fractionation between aqueous Fe(III) and hematite, *Geochim. Cosmochim. Acta* 66 (2002) 2995–3015.
- [26] B.L. Beard, C.M. Johnson, J.L. Skulan, K.H. Nealson, L. Cox, H. Sun, Application of Fe isotopes to tracing the geochemical and biological cycling of Fe, *Chem. Geol.* 195 (2003) 87–117.
- [27] B.L. Beard, C.M. Johnson, J.L. Skulan, L.A. Taylor, H. Sun, L. Cox, K.H. Nealson, M.P. Sinha, M.J. Gerdenich, The Fe-isotope system and its applicability as a biosignature, *Lunar Planet. Sci. Conf. XXXII*, 2001.
- [28] F. Poitrasson, A.N. Halliday, D.-C. Lee, S. Levasseur, N. Teutsch, Origin of the Moon unveiled by its heavy iron isotope composition, *EOS Trans. Suppl.* 83 (47) (2002).
- [29] F. Poitrasson, A.N. Halliday, D.-C. Lee, S. Levasseur, N. Teutsch, Iron isotope evidence for formation of the moon through partial vaporization, *Lunar Planet. Sci. Conf. XXXIV*, 2003.
- [30] R.V. Morris, Origins and size distribution of metallic iron particles in the lunar regolith, *Proc. 11th Lunar Sci. Conf.*, 1980, pp. 1697–1712.
- [31] D.E. Brownlee, B. Bates, L. Schramm, The elemental composition of stony cosmic spherules, *Meteorit. Planet. Sci.* 32 (1997) 157–175.
- [32] D.E. Brownlee, The origin and properties of dust impacting the earth, in: B. Peucker-Ehrenbrink, B. Schmitz (Eds.), *Accretion of Extraterrestrial Matter Throughout Earth's History*, Kluwer Academic, New York, 2001, pp. 1–12.
- [33] X.K. Zhu, Y. Guo, R.K. O'Nions, E.D. Young, R.D. Ash, Isotopic homogeneity of iron in the early solar nebula, *Nature* 412 (2001) 311–313.
- [34] A.W.R. Bevan, P.A. Bland, A.J.T. Jull, Meteorite flux on the Nullarbor region, Australia, in: M.M. Grady, R. Hutchison, G.J.H. McCall, D.A. Rothery (Eds.), *Meteorites: Flux with Time and Impact Effects*, Geological Society, London, 1998.
- [35] T.J. Bernatowicz, R.H. Nichols, C.M. Hohenberg, Origin of amorphous rims on lunar soil grains, *Lunar Planet. Sci. Conf. XXV*, 1994, pp. 105–106.
- [36] B.W. Hapke, W.A. Cassidy, E. Wells, Vapor deposits in the lunar regolith Technical comment, *Science* 264 (1994) 1779.
- [37] T.J. Bernatowicz, R.H. Nichols, C.M. Hohenberg, M. Maurette, Vapor deposits in the lunar regolith Technical comment, *Science* 264 (1994) 1779–1780.
- [38] L.P. Keller, D.S. McKay, Vapor deposits in the lunar regolith-replay, *Science* 264 (1994) 1780.
- [39] J.P. Bibring, J.P. Duraud, L. Durrieu, C. Jouret, M. Maurette, R. Meunier, Ultrathin amorphous coatings on lunar dust grains, *Science* 175 (1972) 753–755.
- [40] W.A. Cassidy, B.W. Hapke, Effects of darkening processes on surfaces of airless bodies, *Icarus* 25 (1975) 371–383.
- [41] B.W. Hapke, W.A. Cassidy, Is the Moon really as smooth as a billiard ball? Remarks concerning recent models of sputter-fractionation on the lunar surface, *Geophys. Res. Lett.* 5 (1978) 297–300.
- [42] D.M. Paruso, W.A. Cassidy, B.W. Hapke, An experimental investigation of fractionation by sputter deposition, *Proc. 9th Lunar Planet. Sci. Conf.*, 1978, pp. 1711–1724.
- [43] R. Christoffersen, L.P. Keller, D.S. McKay, Microstructure, chemistry, and origin of grain rims on ilmenite from the lunar soil finest fraction, *Meteorit. Planet. Sci.* 31 (1996) 835–848.
- [44] J.P. Bradley, Chemically anomalous, preaccretionally irradiated grains in interplanetary dust from comets, *Science* 265 (1994) 925–929.
- [45] W.A. Russell, D.A. Papanastassiou, T.A. Tombrello, The fractionation of Ca isotopes by sputtering, *Radiat. Eff.* 52 (1980) 41–52.
- [46] H.P. Taylor, S. Epstein, O^{18}/O^{16} and Si^{30}/Si^{28} studies of some Apollo 15, 16, and 17 samples, *Proc. 4th Lunar Sci. Conf.*, 1973, pp. 1657–1679.
- [47] G. De Maria, G. Balducci, M. Guido, V. Piacente, Mass spectrometric investigation of the vaporization process of Apollo 12 lunar samples, *Proc. 2nd Lunar Sci. Conf.*, 1971, pp. 1367–1380.
- [48] J. Wang, A.M. Davies, R.N. Clayton, T.K. Mayeda, A. Hashimoto, Chemical and isotopic fractionation during the evaporation of the FeO-MgO-SiO₂-CaO-Al₂O₃-TiO₂ rare earth element system, *Geochim. Cosmochim. Acta* 65 (2001) 479–494.
- [49] L.A. Taylor, Origin of nanophase Fe^o in agglutinates: a radical new concept, in: *The Moon Beyond 2002: Next Steps in Lunar Science and Exploration*, Lunar and Planetary Science Institute, 2002.