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## Trace element geochemistry of Martian iddingsite in the Lafayette meteorite

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**Abstract.** The Lafayette meteorite contains abundant iddingsite, a fine-grained intergrowth of smectite clay, ferrihydrite, and ionic salt minerals. Both the meteorite and iddingsite formed on Mars. Samples of iddingsite, olivine, and augite pyroxene were extracted from Lafayette and analyzed for trace elements by instrumental neutron activation. Our results are comparable to independent analyses by electron and ion microbeam methods. Abundances of most elements in the iddingsite do not covary significantly. The iddingsite is extremely rich in Hg, which is probably terrestrial contamination. For the elements Si, Al, Fe, Mn, Ni, Co, and Zn, the composition of the iddingsite is close to a mixture of ~50% Lafayette olivine + ~40% Lafayette siliceous glass + ~10% water. Concordant behavior among these elements is not compatible with element fractionations between smectite and water, but the hydrous nature and petrographic setting of the iddingsite clearly suggest an aqueous origin. These inferences are both consistent, however, with deposition of the iddingsite originally as a silicate gel, which then crystallized (neoformed) nearly isochemically. The iddingsite contains significantly more magnesium than implied by the model, which may suggest that the altering solutions were rich in  $Mg^{2+}$ .

### Introduction

The Workshop on the Evolution of Martian Volatiles was convened to help resolve problems about distributions, sources, abundances of volatiles on Mars. One approach to these problems is through analyses of Martian volatiles and their products in meteorites; most of the Martian meteorites were affected by water (or other volatiles) on Mars [e.g., Gooding, 1992; Treiman *et al.*, 1993; Romanek *et al.*, 1996]. Ideally, studies of an altered meteorite could provide fluid compositions, fluid-rock ratios, and timings of a volatile-driven process on Mars.

In this work, we continue studies of the Martian alteration materials in Lafayette, one of the nakhlite meteorites. The nakhlites are igneous rocks composed mostly of augite pyroxene and olivine, with many minor phases including silica-rich mesostasis glass [Bunch and Reid, 1975; Berkley *et al.*, 1980; Harvey and McSween, 1992; Treiman *et al.*, 1993]. In the nakhlites, some olivine and mesostasis glass were replaced by iddingsite, an intimate mixture of ferroan smectite clay, ferrihydrite (a poorly crystalline ferric hydroxide hydrate) and small proportions of ionic salt minerals including Mg- and Ca-sulfates, Ca- and Mg-carbonates, and NaCl [Gooding *et al.*, 1991; Treiman *et al.*, 1993]. Iddingsite in the nakhlites is definitely preterrestrial (and therefore Martian) because it is cut by the meteorites' fusion crusts [Gooding *et al.*, 1991; Treiman *et al.*, 1993] and has oxygen isotope abundances consistent with a Martian origin [Karlsson *et al.*, 1992; Romanek *et al.*, 1996].

Lafayette contains the most iddingsite of any nakhlite and so is an ideal target for extended study. To date, Lafayette's iddingsite has been analyzed for bulk composition and

microstructures [Treiman *et al.*, 1993], heavy noble gases [Drake *et al.*, 1994], formation age by  $^{40}\text{Ar}/^{39}\text{Ar}$  [Swindle *et al.*, 1995], oxygen isotopes [Karlsson *et al.*, 1992; Romanek *et al.*, 1996], and hydrogen isotopes [Leshin *et al.*, 1996]. In this paper we report trace element analyses for Lafayette's iddingsite by instrumental neutron activation analysis (INAA) and speculate on the mechanism of iddingsite formation in Lafayette and on the composition of the water that altered Lafayette.

### Sample and Analysis

Fragments of iddingsite, olivine, and pyroxene were removed from 3-5 mm crumbs of Lafayette sample ME2116 from the Field Museum, Chicago, Illinois. Among the crumbs was a range of terrestrial materials, including wood slivers, paint flecks, and blobs of glue. These materials were carefully excluded from the analyzed samples, but could still be sources of contamination. The meteorite has also seen extensive handling in the museum, and could have been exposed to water, saw oils, cleaning solutions, etc. Nonetheless, exception for Hg (see below), no evidence of high and variable abundances of any contaminants was observed.

The crumbs were dissected under a stereo microscope using needles of stainless steel and silica glass; dissected fragments were identified by their colors: orange-brown iddingsite, yellow olivine, and green pyroxene. Iddingsite samples were dissected from two separate crumbs, labeled 9 and 11. Crumb 9 was an exterior piece with fusion crust on one face. All iddingsite from crumb 9 was at least 1 mm distant from the fusion crust, which is probably far enough to ensure minimal movement of volatiles during atmospheric entry [Treiman *et al.*, 1993]. Crumb 11 had no fusion crust and so was probably > 2 mm from the fusion crust. Plagioclase and phosphates appeared white and were avoided or dissected away. Samples for analysis ranged up to ~20  $\mu\text{m}$  in diameter and down to a few microns in diameter.

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Pyroxene grains were clear and glassy, and contained no visible mineral impurities. Olivine grains were also clear and glassy but thin veinlets of iddingsite and minute opaque grains were ubiquitous and could not be dissected out. Iddingsite fragments were brown and essentially opaque, and were commonly intergrown with other silicates. Efforts to clean the iddingsite grains were quite successful, as is shown by consideration of the analytical data below.

Samples for analysis were packaged in sealed high-purity quartz tubes and irradiated for 96 hours at a thermal neutron flux of about  $7.5 \times 10^{13}$  neutrons/cm<sup>2</sup>-sec. After irradiation, the samples were removed from the tubes and weighed into new containers for gamma ray assay. Weights of olivine and pyroxene samples were measured using a Sartorius S3 microbalance to an uncertainty of  $\pm 0.5 \mu\text{g}$ . Olivine sample weights ranged from 5 to 18  $\mu\text{g}$ , and pyroxene sample weights ranged from 14 to 36  $\mu\text{g}$  (Table 1). Samples of iddingsite were too friable for extended sample handling and were not weighed; their weights were calculated after analysis on the assumption that they each contained 29% FeO (anhydrous basis), the average value for iddingsite [Treiman *et al.*, 1993]. Calculated weights for iddingsite samples were between 0.45 and 20  $\mu\text{g}$  (Table 1). The gamma ray counting assay was performed in the

low-background Radiation Counting Laboratory at Johnson Space Center using intrinsic Ge detectors. Analytical standards were glasses used extensively in this lab [Lindstrom *et al.*, 1994] and the low-Ni hexahedrites Lombard and San Martin [Lindstrom and Jones, 1996]. Satisfactory standards are not available for Br, Hg, and Se, so their absolute abundances are based on published cross sections [Mughabghab, 1984] and are probably uncertain to a factor of 2, although differences between samples are much more accurate.

## Analytical Results

Average chemical analyses of "pure" iddingsite, pyroxene, and olivine are given in Table 1, some individual results are graphed in Figures 1 to 3, and the full data set on iddingsite samples is given in Table 2. Our analyses for Na, K, Ca, and Fe compare extremely well with electron microprobe and scanning electron microscope / energy dispersive X ray analyses (SEM/EDX) in the literature [Bunch and Reid, 1975; Boctor *et al.*, 1976; Berkley *et al.*, 1980; Smith *et al.*, 1983; Harvey and McSween, 1992; Treiman *et al.*, 1993]. The olivine and pyroxene in Lafayette are nearly homogeneous in major and minor elements [Berkley *et al.*, 1980; Harvey and McSween, 1992], consistent with the agreement among our analyses and all the literature values.

Rare earth element (REE) concentrations in our samples can be compared to the ion microprobe (SIMS) analyses Lafayette minerals of Wadhwa and Crozaz [1995]. REE in olivine, pyroxene, and iddingsite are low, and both the SIMS and INAA techniques have difficulties with some elements in some samples. Neither SIMS nor INAA can analyze adequately for light REE (LREE) in olivine. We obtained  $250 \pm 20$  ppb Yb and  $56 \pm 10$  ppb Lu in five grains of olivine, in reasonable agreement with a single value from Wadhwa and Crozaz [1995] of  $284 \pm 24$  ppb Yb. Figure 3a shows that the lowest eight of our 16 analyses for La and Sm in pyroxenes fall near the middle of the range quoted by Wadhwa and Crozaz [1995], while our Yb results (which we believe are accurate within the quoted uncertainties) suggest that SIMS analyses for Yb in pyroxene are overestimated by ~20%. Nonetheless, when viewed on the usual REE plot (Figure 4), the agreement is good.

Before considering the geochemistry of the analyzed samples, it is important to discuss artifacts that might arise from uncertainties in sample masses, sample impurities in the form of mineral admixtures, and possible terrestrial contamination.

Even with the use of a high-quality microbalance, the smaller sample masses are significantly uncertain. For the pyroxenes and olivines, weighing uncertainties would shift the points a few percent along lines through the origin in Figure 1. This effect alone cannot explain the range of analyses, which must reflect grain-to-grain compositional variability in these minerals [Berkley *et al.*, 1980; Harvey and McSween, 1992; Wadhwa and Crozaz, 1995]. Also note that most of the spread in Cr in the pyroxenes is perpendicular to the trend expected from weighing errors.

Because we were unable to weigh the iddingsite samples, their analyses have been normalized to the average of 29.0% FeO determined by Treiman *et al.* [1993]. That paper gave analyses of seven petrographically different iddingsite types whose FeO contents had a standard deviation of only 5%, so a similar variability in our iddingsite samples could lead to small displacements along lines through the origin in Figure 1. Even much larger changes in the apparent weights could not explain

**Table 1.** Average Chemical Compositions by INAA

Element	Iddingsite	Pyroxene	Olivine
Na <sub>2</sub> O %	0.30 $\pm$ 0.07	0.245 $\pm$ 0.016	—
K <sub>2</sub> O %	0.7 $\pm$ 0.3	<0.02	—
CaO %	2 $\pm$ 1	19.3 $\pm$ 1.7	<1
FeO %	$\approx$ 29.0	14.4 $\pm$ 0.7	50.8 $\pm$ 1.9
Sc ppm	0.4 $\pm$ 0.2	69 $\pm$ 3	8.5 $\pm$ 0.6
Cr ppm	5 $\pm$ 2	2630 $\pm$ 160	30 $\pm$ 10
Co ppm	55 $\pm$ 5	37.5 $\pm$ 0.9	111 $\pm$ 5
Zn ppm	160 $\pm$ 50	85 $\pm$ 26	190 $\pm$ 20
Rb ppm	40 $\pm$ 20	<10	<30
Cs ppm	20 $\pm$ 10	<0.7	<0.7
Sr ppm	250 $\pm$ 100	<160	<200
Ba ppm	150 $\pm$ 80	<60	<60
La ppm	<0.1	0.34 $\pm$ 0.08	<0.1
Ce ppm	<1	1.6 $\pm$ 0.3	<0.8
Sm ppm	0.04 $\pm$ 0.01	0.54 $\pm$ 0.04	<0.03
Eu ppm	<0.2	<0.3	<0.14
Tb ppm	<0.2	<0.2	<0.15
Yb ppm	<0.2	0.31 $\pm$ 0.02	0.25 $\pm$ 0.03
Lu ppm	<0.03	0.052 $\pm$ 0.008	0.056 $\pm$ 0.010
Hf ppm	<0.3	<0.3	<0.5
U ppm	<0.2	<0.5	<0.6
Th ppm	<0.2	<0.2	<0.2
As ppm	1.0 $\pm$ 0.4	<0.2	<0.7
Sb ppm	<0.3	<0.06	<0.1
W ppm	1.0 $\pm$ 0.5	<0.5	<1.2
Ir ppb	<8	10 $\pm$ 2	<10
Au ppb	<8	<4	<5
Br ppm	9 $\pm$ 3	<0.7	<2
Hg ppm	150 $\pm$ 80	<2	<6

Iddingsite analyses are pegged to FeO=29%, the average value (anhydrous basis) from Treiman *et al.* [1993], and uncertainties are primarily estimated from the scatter in plots such as Figure 6. See Table 2 for sample masses and details. Pyroxene analysis is mass-weighted average of samples Px1 (28.8  $\mu\text{g}$ ), Px2a (28.8  $\mu\text{g}$ ), Px4a (20.8  $\mu\text{g}$ ), and Px5b (27.1  $\mu\text{g}$ ). Olivine analysis is mass weighted average of samples Ol2a (17.9  $\mu\text{g}$ ), Ol2c (11.4  $\mu\text{g}$ ), and Ol3a (9.9  $\mu\text{g}$ ). Uncertainties for pyroxene and olivine are 1  $\sigma$  of analysis or estimated 1  $\sigma$  of mean, whichever is greater. All upper limits are 2  $\sigma$ .

many of the compositional differences observed. For example, consider the iddingsite sample with the highest Sc content (Figure 2d). It is tempting to suppose that this sample actually had slightly lower FeO (~25%) and fell on the dotted mixing line between iddingsite and pyroxene, but this effect alone would not explain its high Cr content (Figure 2b).

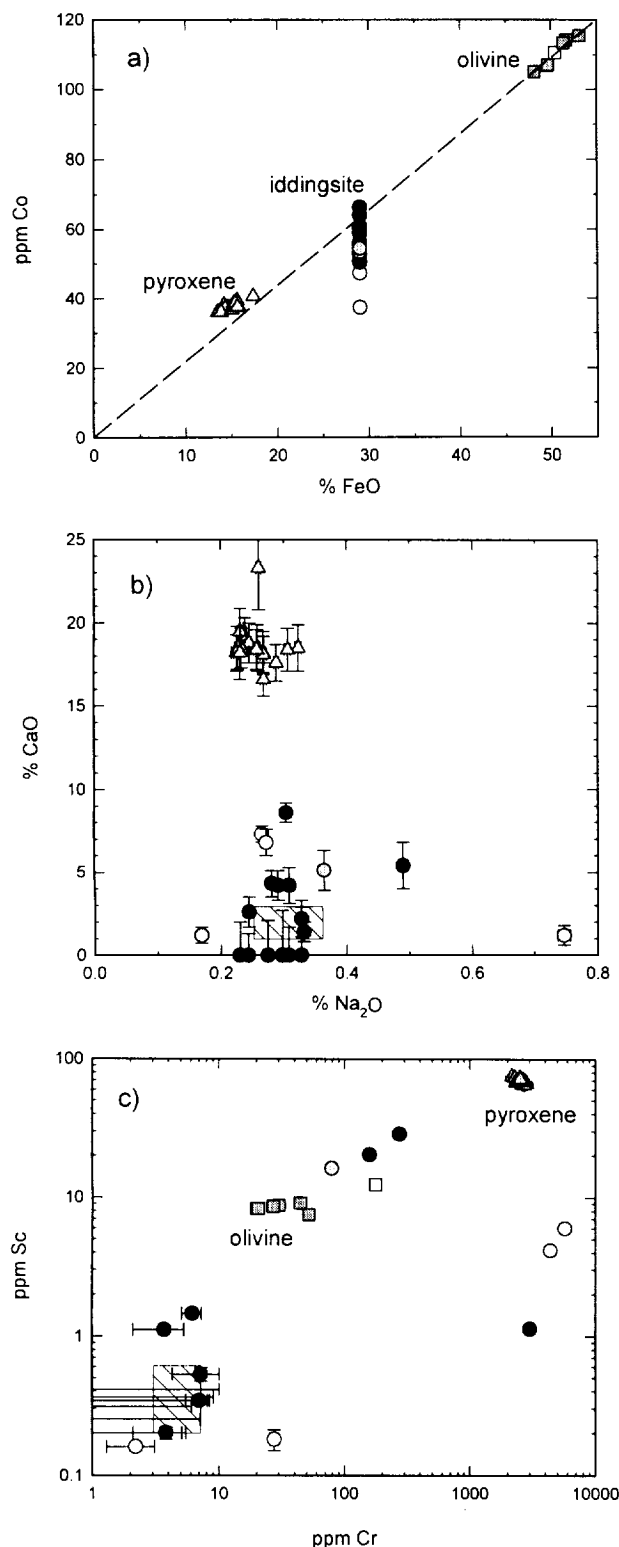
Sample purity could not be completely assured by visual inspection, so we used the analyses themselves as indications of

mineral admixtures. Mixing lines on Figure 2 can be used to set upper limits on the amounts of contamination of one mineral with another. Chromium abundances show that most iddingsite samples contain negligible titanomagnetite, the most Cr-rich oxide phase found in Lafayette [Boctor *et al.*, 1976]; a few samples may contain ~6-12% titanomagnetite by weight. The same Cr data show that 12 of the 19 iddingsite samples contain <0.5% pyroxene (Figure 2c), in agreement with Sc data (Figure 2f). Estimating the amounts of olivine in the iddingsite samples is more difficult, but Sc data show that none are highly contaminated and most can contain no more than 5% olivine. The telltales for admixture of phosphate or mesostasis material were high abundances of the LREE and/or a La/Sm abundance ratio significantly above normal for the host phase. Impurity pyroxene in the iddingsite was apparent by moderately elevated Sc, Cr, and REE abundances (especially Sm), and a La/Sm ratio significantly below chondritic. With these criteria, we found that some samples of iddingsite contained detectable admixtures of other phases, and that most pyroxene and olivine samples were pure or nearly so.

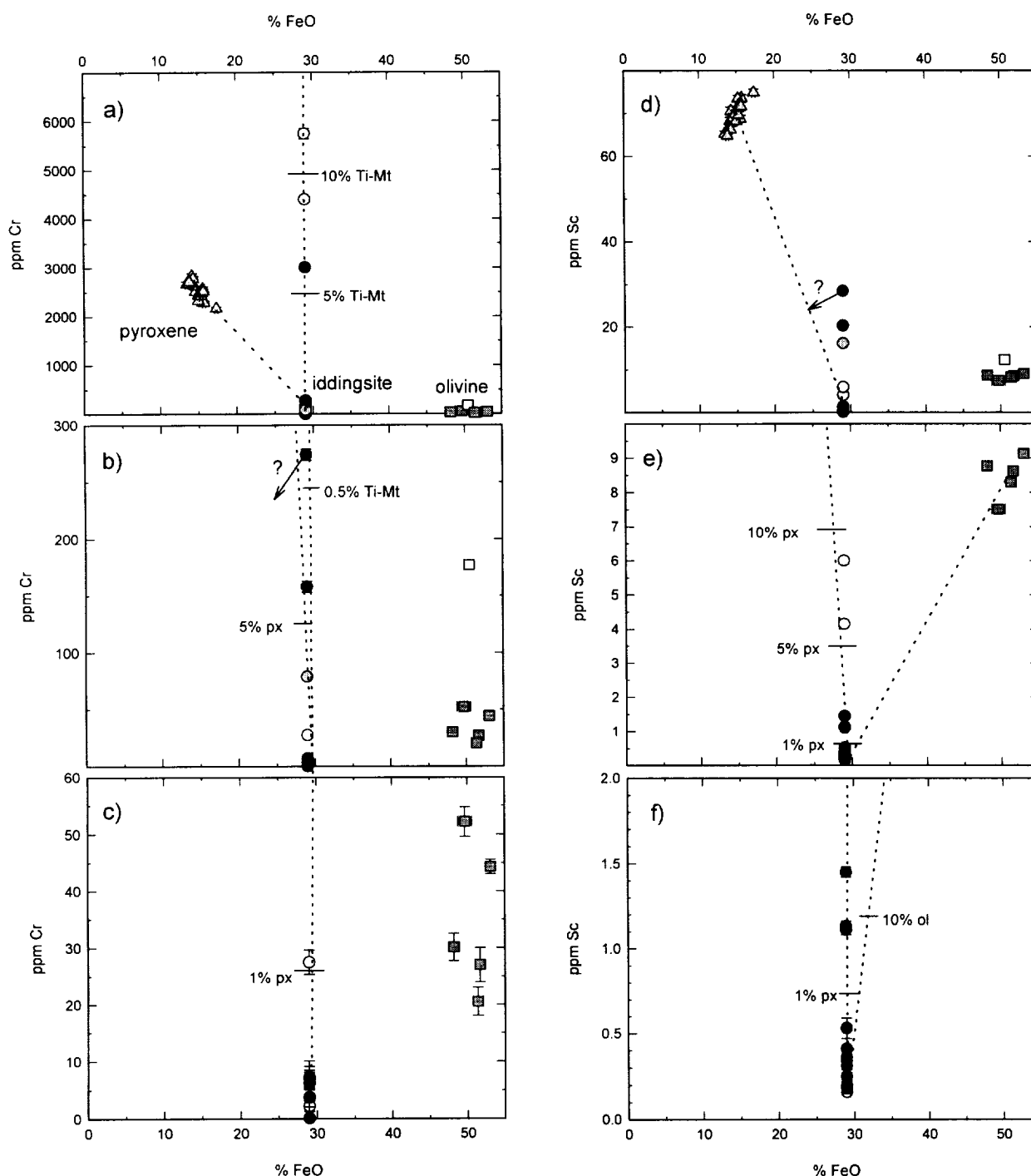
## Mercury

One advantage of INAA is that gamma-ray spectra record the presence of all favorable neutron-capture isotopes regardless of whether their presence is anticipated. The discovery of very high levels of mercury in these iddingsite samples was certainly a surprise. Because of a lack of a standard for Hg, absolute values are uncertain, but are clearly in the range of hundreds of ppm. To our knowledge, bulk samples of Lafayette have not been analyzed for Hg, but the compositionally similar Nakhla has only 0.23 ppm Hg [Ehmann and Lovering, 1967]. The smallest iddingsite samples have the highest Hg contents, and none of the six samples weighing more than 3  $\mu$ g contains more than 100 ppm Hg (Figure 5a). Much lower amounts of Hg (~10 ppm or less) are observed in some of the pyroxene and olivine grains.

Most likely, the high Hg in the Lafayette samples represents terrestrial contamination. As mentioned above, our samples were obtained from meteorite crumbs from the bottom of a standard museum box, where they probably resided for many years. We hypothesize that at some time the sample was



**Figure 1.** Results of INAA of minerals from Lafayette. For these and subsequent graphs, triangles are pyroxenes, squares are olivines, and circles are iddingsite samples. Error bars ( $1\sigma$ ) are based mostly on counting statistics, and  $2\sigma$  upper limits are shown for elements below detection limits. Among the iddingsites, several samples have distinctive chemical characteristics, and are shown as open or shaded circles, while the majority are shown as solid circles. (a) Co/Fe ratios are nearly the same for all three phases. Concentrations are based on actual weights for olivine and pyroxene samples but are calculated assuming FeO=29.0% for the iddingsites. (b) Na<sub>2</sub>O abundances are similar for all of the pyroxenes and most of the iddingsite samples, but CaO contents of the iddingsites vary by 3-4x. Estimates of best values are shown by the hatched box and are given in Table 1. (c) Variations in Sc and Cr can be sizable (note log scales). Some of the iddingsite samples actually may be mixtures with olivine or pyroxene (but keep in mind that mixing lines are not straight on log-log plots), while others clearly are not.



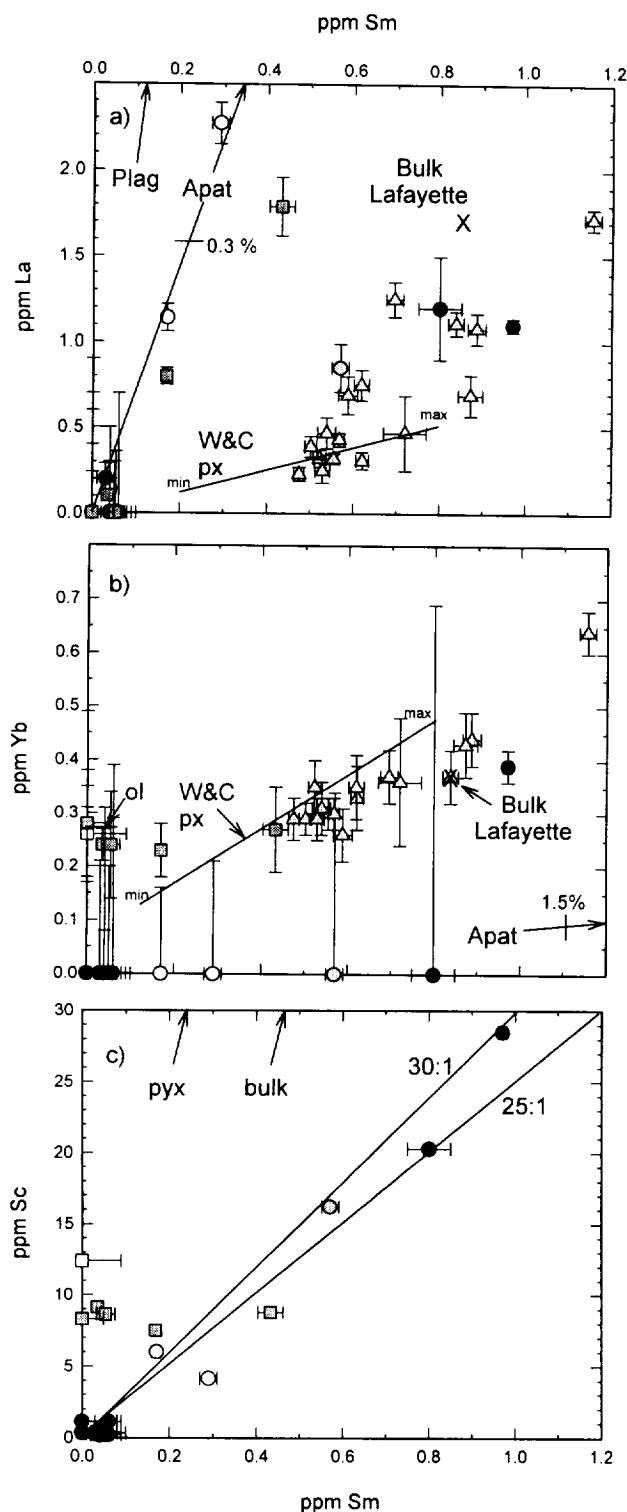
**Figure 2.** Fe-Cr-Sc systematics. These linear plots show the effects of mixing of other minerals with the separated iddingsite samples (dotted lines). If the FeO contents of individual iddingsite samples differ from the assumed value of 29.0% FeO, the points would shift along lines through the origins of the plots. See text for discussion.

exposed to Hg vapor which was adsorbed on the surfaces of the grains, especially on the iddingsite, which because of its fibrous character, has a larger effective surface area. The Hg is particularly obvious on the smallest samples because of their greater surface per unit mass. We consider the fact that all iddingsite samples are rich in Hg to some degree to argue strongly for a vapor phase contamination, since particulates would be expected to produce a much wider range of apparent

Hg concentrations (200 ppm Hg in a 1  $\mu$ g sample corresponds to a sphere of Hg only 2.5  $\mu$ m in diameter).

### Iddingsite Element Systematics

Our analyses for Na, K, Ca, and Fe in Lafayette's iddingsite are quite comparable to electron microprobe and SEM/EDX analyses in the literature (Table 3) [Bunch and Reid, 1975;



**Figure 3.** Rare earths in Lafayette minerals. INAA results from this work are compared with SIMS analyses of *Wadhwa and Crozaz* [1995]. Arrows point toward their analyses of chlorapatite and plagioclase, and the solid line connects their minimum and maximum abundances observed in pyroxenes. One iddingsite samples is off scale, near the chlorapatite mixing line at 2% (La=12.5, Sm=1.4 ppm). Also shown is the bulk composition of Lafayette [*Schmitt and Smith, 1963*].

*Boctor et al., 1976; Treiman et al., 1993*]. The most significant difference between our results and literature values is for CaO. Electron microbeam analyses of iddingsite in the nakhlites are varied, but rarely if ever contain more than 2% CaO (vis. Table 3 and graphs in works by *Gooding et al. [1991]* and *Treiman et al. [1993]*). Six of our upper limits are consistent with this value, but many of our samples have considerably more CaO (Figure 1b, Table 2). The high CaO observed here probably reflects the presence of calcium-rich salt phases like calcite [*Treiman et al., 1993; Gooding et al., 1991, Figure 5d*]. Larger samples would be more likely to include grains of Ca-rich salts, which could be avoided in electron microbeam analyses.

From our INAA data, the strongest compositional signature of the iddingsite is its enrichment in alkalis: K<sub>2</sub>O ranges from 0.36 to 1.3% and Cs from 4 to 36 ppm (Figure 6). Rb is also enriched, but the INAA data are much less precise (Figure 6a). All of the Cs in the rock (0.3 ppm, [*Treiman et al., 1986*]) could be accounted for by 1.5% of the rock being iddingsite with 20 ppm Cs. The iddingsite is also enriched in Br, with most Br concentrations scattering near 10 ppm (Figure 6c); these results may be subject to ~2x systematic errors, since no Br standard was analyzed with the samples.

Other elements detected with reasonable accuracy in the iddingsite include Zn, which varies from about 80 to 280 ppm with no apparent pattern (Figure 6b), and Co, which varies from 40 to 68 ppm. This variation in Co abundance is more than would be expected, considering the high accuracy of the analyses and the normalization to FeO. Although the uncertainties in their analyses are much greater, As (Figure 6f) and W are observed in most of the iddingsite samples, with the remainder giving only upper limits in agreement with the averages of about 1 ppm for both elements. Sr and Ba are observed at about 300 and 150 ppm, respectively, in the largest samples.

A detailed examination of trace element covariations in our analyses of Lafayette's reveals mineral admixtures, but few systematic variations ascribable to the iddingsite itself (Figures 1 and 6). Iddingsite also shows significant, but random, variations in major element composition [*Treiman et al., 1993*]. Abundances of most elements in individual samples appear to be randomly distributed, with variances well beyond the analytical error limits. The variations in most elemental abundances appear to be effectively uncorrelated with those of other elements (save correlations clearly arising from mineral admixtures).

Most of the iddingsite samples have REE abundances too low for accurate analysis by INAA. Sensitivities for Sm by INAA range from a few ppb to about 10 ppb in the smallest samples. The best low number obtained was  $36 \pm 3$  ppb, or about  $0.24 \times \text{CI}$ , and 13 of the other iddingsite particles have Sm contents or upper limits consistent with this value. Abundances of other REE in these low-REE samples were also low,  $<0.5 \times \text{CI}$ . The remaining six iddingsite particles have considerably higher REE contents. The REE in three particles can be readily explained by admixture of 0.2-2.0% chlorapatite (SIMS analyses by *Wadhwa and Crozaz* [1995], Figure 3a), as can several of the olivines and pyroxene samples that are displaced toward higher REE and, especially, higher LREE. The remaining three iddingsite particles scatter in with the more REE-rich pyroxenes on the La versus Sm plot (Figure 3a). These particles are also enriched in

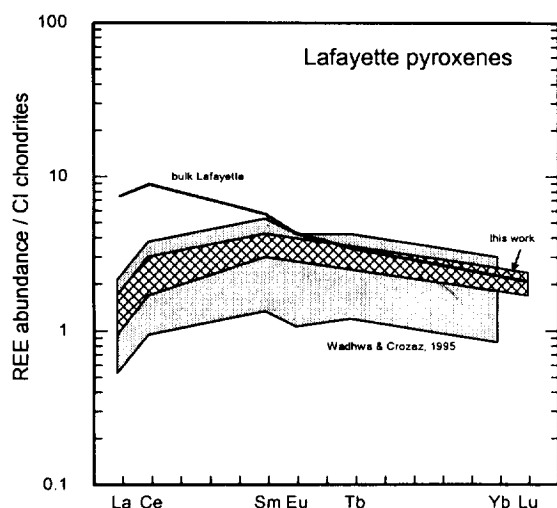
**Table 2.** Compositions of Iddingsite Samples from Lafayette

	Idd-9A	Idd-9B	Idd-9C	Idd-9D	Idd-9E	Idd-9F	Idd-9G	Idd-9H	Idd-9I	Idd-9J
$\mu\text{g}$	19.4	19.3	10.7	4.44	2.70	2.12	1.33	1.23	2.19	0.70
Na <sub>2</sub> O	0.265±0.003	0.304±0.004	0.273±0.004	0.331±0.005	0.327±0.005	0.243±0.004	0.290±0.004	0.231±0.003	0.297±0.004	0.308±0.004
K <sub>2</sub> O	0.380±0.008	0.366±0.007	0.432±0.016	0.833±0.030	0.94±0.04	0.721±0.026	0.562±0.026	0.420±0.021	0.67±0.03	0.544±0.024
CaO	7.3±0.5	8.6±0.6	6.7±0.8	1.4±0.6	<3.0	<1.3	4.2±0.9	<2.0	<2.7	4.3±1.0
FeO	29.0±0.4	29.0±0.4	29.0±0.4	29.0±0.3	29.0±0.3	29.0±0.3	29.0±0.3	29.0±0.3	29.0±0.3	29.0±0.4
Sc	0.160±0.014	28.5±0.3	0.184±0.026	1.445±0.029	1.11±0.03	0.338±0.022	0.200±0.020	0.314±0.020	1.13±0.03	0.35±0.05
Cr	2.2±0.9	274±4	27.6±2.1	6.2±1.0	3.7±1.6	7.0±1.4	3.8±1.6	<5.6	3010±40	<8
Co	47.2±0.8	55.6±0.9	51.0±1.0	54.4±0.8	64.0±0.9	58.8±0.9	52.4±0.8	50.2±0.8	60.1±0.9	52.6±1.4
Zn	88±13	148±14	98±18	182±10	150±11	145±12	108±10	140±10	197±13	125±26
Rb	26±9	19±9	26±11	<45	49±11	36±16	<60	<52	35±16	<70
Cs	17.4±0.8	13.7±0.8	19.2±0.9	12.0±0.4	26.7±0.8	11.7±0.5	20.3±0.6	21.1±0.6	19.3±0.6	24.5±1.5
Sr	340±90	<360	340±140	<230	<250	<390	270±100	240±80	<340	<900
Ba	110±40	110±50	220±70	<230	<290	<260	<270	<330	<240	<340
La	0.14±0.02	1.10±0.04	<0.22	12.2±0.3	<0.6	<0.6	<0.7	<0.9	<0.7	<0.7
Ce	<2.0	7.4±1.1	3.0±0.9	22.5±0.9	<2.3	<2.8	<1.3	<1.7	<2.7	<4.8
Sm	0.036±0.003	0.971±0.013	0.042±0.015	1.37±0.04	0.055±0.027	<0.08	0.063±0.029	<0.10	<0.08	<0.09
Eu	<0.17	0.37±0.16	<0.31	0.34±0.05	0.15±0.06	0.20±0.09	<0.17	0.21±0.06	<0.29	<0.9
Tb	<0.21	<0.38	<0.24	<0.25	<0.34	<0.24	<0.21	<0.17	<0.4	<0.5
Yb	<0.07	0.39±0.03	<0.20	0.27±0.09	<0.4	<0.32	<0.29	<0.27	<0.29	<0.4
Lu	<0.023	0.061±0.007	<0.033	0.045±0.016	<0.08	<0.05	<0.06	<0.05	<0.06	<0.07
Hf	<0.4	0.51±0.23	<0.3	<0.28	<0.3	<0.4	<0.3	<0.3	<0.5	<0.7
Ta	<0.8	<0.8	<0.9	<0.6	<1.0	<1.0	<0.9	<0.8	<1.5	<1.8
U	<0.5	<0.14	<2.0	<3.4	<4	<2.3	<3	<2.9	<0.24	<2.8
Th	<0.19	<0.44	<0.42	0.89±0.12	<0.31	<0.32	<0.3	<0.27	<0.4	<0.4
As	0.75±0.06	0.52±0.07	1.09±0.24	<1.5	<1.6	0.9±0.3	1.3±0.4	1.8±0.4	<1.2	1.1±0.4
Sb	0.15±0.03	0.11±0.04	<0.30	<0.33	<0.7	<0.6	<0.4	<0.5	<0.6	<0.6
W	0.84±0.07	0.50±0.08	1.11±0.25	<1.6	<2.0	<1.5	2.2±0.5	3.3±0.5	1.6±0.6	1.3±0.6
Ir	<0.006	<0.008	<0.010	<0.007	<0.008	<0.009	<0.007	<0.008	<0.019	<0.017
Au	0.006±0.001	<0.005	<0.015	<0.007	<0.042	<0.029	<0.031	<0.034	<0.032	<0.028
Br	11.5±0.	8.5±0.3	11.5±0.9	9.0±1.4	8.8±1.7	7.5±1.3	10.0±1.6	16.0±1.7	9.5±1.9	12.8±1.4
Hg	65.6±1.5	42.1±1.5	75.3±3.0	93.3±2.1	292±4	212±4	129.5±2.9	69.6±2.2	163±3	117±5

	Idd-9K	Idd-9L	Idd-9M	Idd-9N	Idd-9O	Idd-9P	Idd-11A	Idd-11B	Lafay-11C
$\mu\text{g}$	1.35	0.98	0.56	0.63	0.47	0.74	7.65	1.60	6.69
Na <sub>2</sub> O	0.489±0.007	0.273±0.004	0.307±0.004	0.245±0.004	0.327±0.005	0.280±0.004	0.169±0.003	0.364±0.005	0.747±0.010
K <sub>2</sub> O	0.77±0.03	0.75±0.03	0.94±0.04	0.555±0.021	0.678±0.027	0.532±0.029	0.582±0.022	1.05±0.04	0.408±0.014
CaO	5.4±1.4	<2.1	<1.6	2.6±0.9	2.2±1.0	4.2±0.8	1.1±0.5	5.1±1.1	1.2±0.6
FeO	29.0±0.4	29.1±0.3	29.1±0.4	29.0±0.4	29.0±0.4	29.0±0.4	29.0±0.3	29.0±0.3	29.0±0.4
Sc	20.35±0.25	0.249±0.021	0.35±0.03	0.54±0.06	0.41±0.06	0.20±0.04	4.15±0.05	16.23±0.19	5.99±0.09
Cr	157±4	<7	<8	7.2±2.9	<9	<5	4400±60	78±3	5740±100
Co	56.2±1.0	60.8±1.0	66.4±1.2	53.4±1.5	59.2±1.7	52.7±1.3	53.3±0.7	54.4±0.9	37.3±0.9
Zn	166±17	150±13	177±19	103±28	130±30	137±25	259±9	206±16	285±24
Rb	<70	<80	<100	51±23	<100	<60	<34	<60	<28
Cs	15.6±0.8	14.0±0.6	34.1±1.1	13.3±1.2	32.0±2.0	21.0±1.4	10.2±0.3	18.2±0.8	4.2±0.7
Sr	<400	<350	<600	<1700	<380	<400	<210	<500	<400
Ba	<500	<380	<390	<330	<410	<380	<180	<340	<170
La	1.20±0.27	0.21±0.09	<0.7	<0.35	<0.4	<0.30	2.27±0.12	0.85±0.14	1.14±0.08
Ce	3.1±1.4	1.7±0.7	<2.8	<6	<4	<3.5	5.6±1.0	2.8±1.2	<4.0
Sm	0.79±0.05	0.033±0.016	<0.07	0.041±0.013	0.044±0.018	0.049±0.012	0.294±0.017	0.570±0.024	0.169±0.010
Eu	<0.6	0.30±0.11	<0.37	<0.5	<0.5	<0.4	0.10±0.04	0.42±0.18	<0.54
Tb	<0.6	<0.25	<0.3	<0.6	<0.6	<0.4	<0.21	<0.5	<0.5
Yb	<0.7	<0.22	<0.5	<0.31	<0.28	<0.20	<0.21	<0.34	<0.16
Lu	<0.13	<0.05	<0.07	<0.06	<0.10	<0.06	<0.04	<0.07	<0.029
Hf	<1.1	<0.5	<0.6	<1.1	<1.0	<0.6	<0.41	<0.8	<1.0
Ta	<3.0	<1.4	<2.1	<1.4	<1.8	<1.6	1.44±0.28	<1.9	2.1±0.7
U	<3	<2.1	<4	<1.9	<3.2	<1.1	<1.1	<1.8	<0.9
Th	<0.7	<0.5	<0.6	<0.7	<0.7	<0.5	<0.31	<0.5	<0.6
As	<1.4	0.9±0.3	1.1±0.5	0.99±0.28	1.5±0.3	1.37±0.26	0.31±0.15	0.74±0.23	0.25±0.12
Sb	<0.7	<0.5	<0.6	<0.6	<0.7	<0.24	<0.24	<0.29	<0.22
W	1.8±0.6	<0.9	<2.4	1.05±0.28	0.8±0.4	2.9±0.4	2.34±0.25	<1.4	2.58±0.21
Ir	<0.014	<0.013	<0.019	<0.024	<0.023	<0.015	0.0131±0.002	<0.013	0.016±0.006
Au	<0.037	<0.017	<0.023	<0.020	<0.025	<0.020	<0.011	<0.014	<0.008
Br	8.9±1.7	4.7±1.1	8.7±1.5	8.9±1.0	10.8±1.2	11.8±1.0	1.8±0.8	3.6±0.8	3.0±0.5
Hg	64±3	198±3	347±6	129±5	244±7	173±5	13.9±1.2	12.6±1.8	8.4±1.9

Abundances in ppm, except oxides in %. Uncertainties are 1  $\sigma$ ; upper limits are 2  $\sigma$ . Masses are in micrograms.

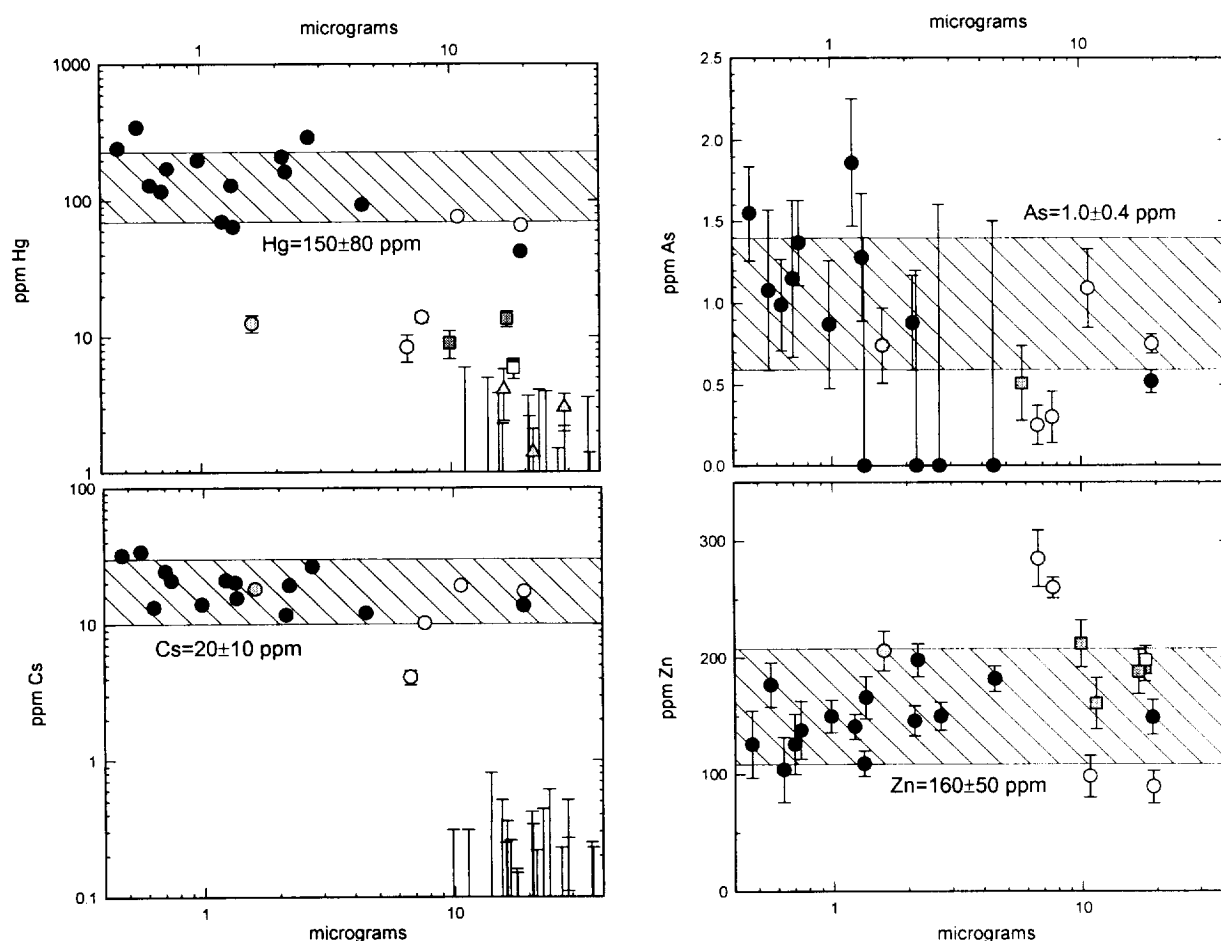


**Figure 4.** Rare earth patterns of Lafayette pyroxenes. INAA results from this work (hachured) are compared with SIMS analyses of Wadhwa and Crozaz [1995] (solid fill). Also shown is the pattern for a bulk sample of Lafayette [Schmitt and Smith, 1963].

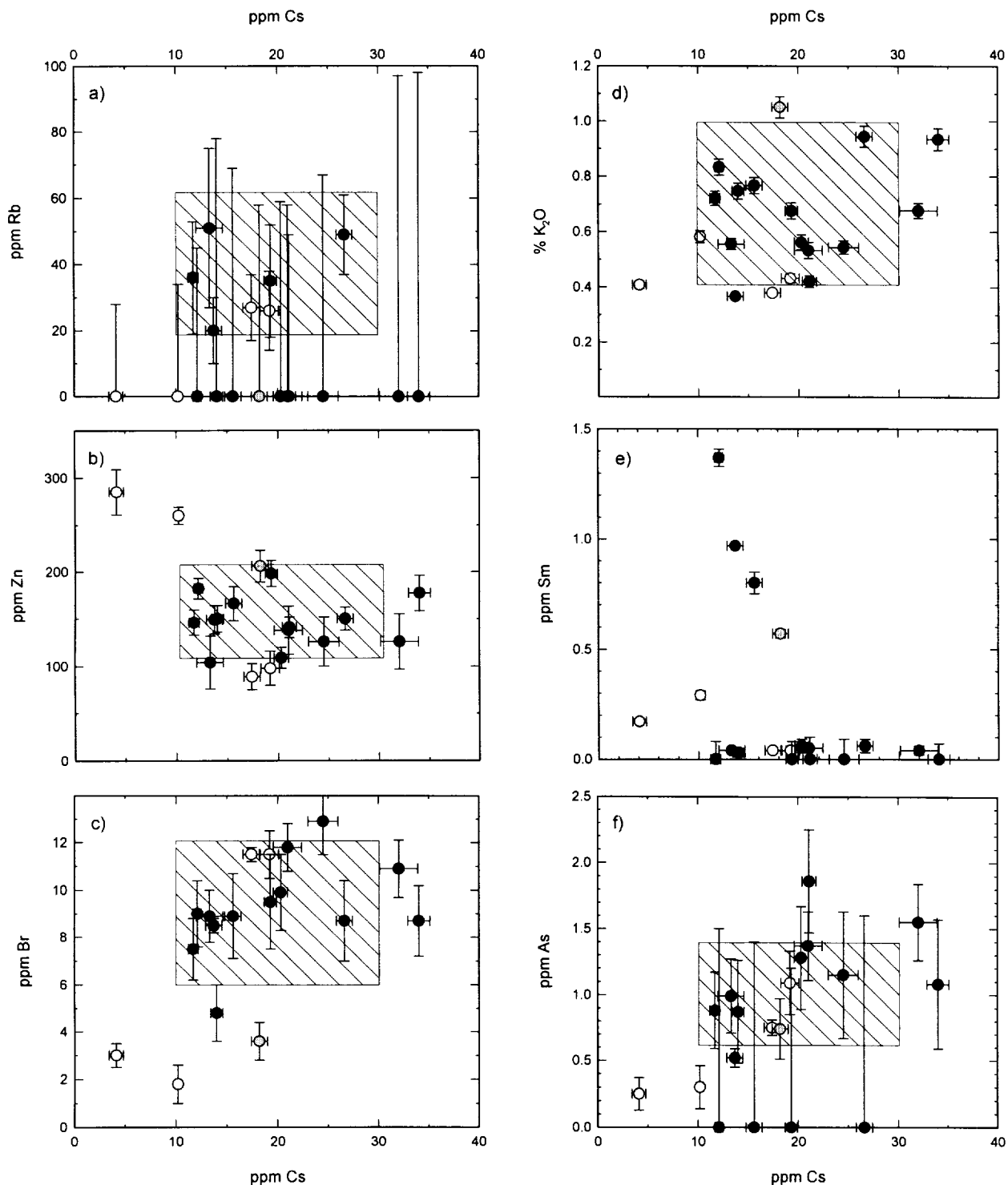
Sc and fall at the Sc-rich end of the trend on Figure 1c. These particles also all have a Sc:Sm ratio of about 25-30:1 (Figure 3c), suggesting that Lafayette contains a minor phase with Sc:Sm in that proportion.

### Iddingsite Formation: A Chemical Model

From major and minor element analyses, Treiman *et al.* [1993] proposed a simple model for formation of Lafayette's iddingsite: all major elements were mobile, at least to some extent; the major element composition of the iddingsite was controlled by the rock; and the composition of the iddingsite is approximated by 50% Lafayette olivine + 40% Lafayette mesostasis glass + 10% water, the 50-40-10 model. For analyses given on an anhydrous basis, as in the 29% FeO of Tables 1, 3, and 4, this model corresponds approximately to 55% olivine + 45% mesostasis glass (the 55-45 anhydrous model). The model was consistent with abundances of Fe, Mn, Si, and Al in the iddingsite [Treiman *et al.*, 1993], predicted significantly more Na and K than were analyzed, and predicted about 60% less Mg than was analyzed (Table 3). In general terms, the model implies that olivine and mesostasis glass dissolved quantitatively into solution, and that the iddingsite was precipitated from solution with limited chemical fractionation.



**Figure 5.** Trace element concentrations of Lafayette samples as functions of sample size. Except for Hg, there are no apparent variations of trace element content with sample size. Dashed horizontal lines are estimated "best values" given in Table 1. Note that all of the iddingsite samples have Cs abundances > 4 ppm, but none of the other minerals are above 1 ppm.



**Figure 6.** Attempts to correlate other elements with Cs. (a) Uncertainties are high for Rb analyses, but all values and upper limits are in agreement with the estimate of  $40 \pm 20$  ppm Rb, and no clear correlation with Cs. (b) Except for a few outliers, Zn concentrations are fairly constant at  $150 \pm 50$  ppm. (c) Br values, which are subject to possible systematic errors (see text), are about  $9 \pm 3$  ppm except for a few outliers. The two samples with lowest Br and Cs, which seem to suggest a correlation between Br and Cs, are also anomalous in containing high Zn and Sm abundances. (d) Potassium values scatter near  $0.75 \pm 0.15\%$  K<sub>2</sub>O but are essentially not correlated with Cs values. (e) Sm does not correlate with Cs, with most samples having  $< 0.1$  ppm Sm, but six are much higher. (f) Arsenic values are mostly  $1.0 \pm 0.5$  ppm and lack a strong correlation with Cs.



**Table 3.** Electron Microbeam Analyses of Iddingsite From the Lafayette and Nakhla Meteorites

	Lafayette			Nakhla	
	<i>Treiman et al.</i> [1993]	<i>Boctor et al.</i> [1976]	<i>Bunch and Reid</i> [1975]	<i>Gooding et al.</i> [1991]	<i>Bunch and Reid</i> [1975]
SiO <sub>2</sub>	49.1	50.0	49.2	44.62	49.2
TiO <sub>2</sub>	0.01	<0.01	—	0.02	—
Al <sub>2</sub> O <sub>3</sub>	4.77	4.87	6.31	0.82	6.54
Cr <sub>2</sub> O <sub>3</sub>	0.00	0.03	—	0.03	—
FeO	29.01	30.66	32.4	42.06	31.0
NiO	0.10	—	—	<0.01	—
MnO	0.48	—	0.32	0.70	0.42
MgO	13.90	11.7	9.98	7.57	10.6
CaO	0.70	1.54	0.75	1.27	0.82
Na <sub>2</sub> O	0.30	0.19	0.15	1.29*	0.24
K <sub>2</sub> O	0.41	0.94	0.92	0.67	1.24
P <sub>2</sub> O <sub>5</sub>	0.09	—	—	0.07	—
SO <sub>3</sub>	0.69	—	—	0.15	—

All analyses normalized to 100% on elements analyzed. Note the variable compositions, and that the Ca abundances are uniformly lower than to our INAA data (Table 1).

\* Suspected to be erroneous.

Using our new data and selected older data, we can test and modify the model of *Treiman et al.* [1993] but only for elements where we know or can reasonably infer abundances in the mesostasis glass. Lafayette's mesostasis glass has been analyzed previously for K and Na and we assume that the glass contains negligible concentrations of Zn, Ni, Co, Sc, and Cr.

#### Ferromagnesian Elements

Abundances of Co, Zn, and Ni are adequately predicted by the 55-45 (anhydrous) model [*Treiman et al.*, 1993], following the behavior of Fe and Mn. Lafayette iddingsite contains  $55 \pm 5$  ppm Co, approximately 45-54% of the 111 ppm Co in Lafayette's olivine (Table 1). This percentage is within uncertainty and sample heterogeneity of the model value of 55%. Lafayette iddingsite contains about  $160 \pm 50$  ppm Zn, between 58 and 110% of the 190 ppm Zn in Lafayette's olivine (Table 1). The model predicts 55%. Finally, Lafayette iddingsite contains 0.10% NiO, approximately 60% of the 0.16% in the olivine [*Treiman et al.*, 1993], and consistent with the 55-45 anhydrous model (Table 4).

#### Trivalent Compatible Elements

The heavy REE, Sc, and Cr do not fit within the model. The critical data here are that these elements are measurably abundant in Lafayette olivine but are undetectable or at very low levels in the iddingsite (Table 1). In fact, the "pure" iddingsite contains 2% of the Sc abundance in olivine, 7% of the Cr abundance in olivine, and <33% of the Yb and Lu abundances in the olivine. It is difficult to tell whether these elements were immobile during alteration or whether the small proportion of them in the reacted olivine was flushed out of the rock.

#### Alkali Metals

Abundances of Na and K in the iddingsite are not consistent with the 50-40-10 model. From the 1.7% Na<sub>2</sub>O in the mesostasis glass, the model would predict ~0.7% Na<sub>2</sub>O in the iddingsite,

while it actually contains ~0.3% (Table 4). The abundance of K in the mesostasis glass is quite variable, with K<sub>2</sub>O values ranging from 0.5% to 10% [*Berkley et al.*, 1980; *Treiman et al.*, 1993]. Abundances of K span consistency with the model; the iddingsite contains only 5% of the K in the high-K mesostasis glass analysis of *Treiman et al.* [1993] and 80% of the K in the low-K analysis, compared to the predicted 45%. The predicted iddingsite composition of Table 4 uses the average analysis value from *Berkley et al.* [1980], and yields a K<sub>2</sub>O abundance 7.5 times the analyzed value.

#### An Expanded Chemical Model

The "50% olivine + 40% mesostasis glass + 10% water" model of iddingsite formation was shown to be consistent with abundances of Si, Fe, Mn, and Al [*Treiman et al.*, 1993]. From the data here, abundances of the ferromagnesian cations Co, Ni, and Zn are also passably consistent with the model. Of ferromagnesian elements, only Mg deviates from the model in being significantly more abundant in the iddingsite than predicted. Other elements, where data are available, are significantly less abundant in the iddingsite than the model would predict. It is possible that many elements, particularly trivalent and higher valency cations, were not mobile during iddingsite formation and thus could not follow the model. It is equally possible that some elements, like the alkalis, might have been dissolved quantitatively from the mesostasis glass but not retained quantitatively in the iddingsite.

#### Iddingsite Formation: A Physicochemical Mechanism

An important goal of studying the aqueous alteration products in the Martian meteorites is to characterize the fluids that produced them. Lafayette's iddingsite is critically important in this effort, as it is the most abundant aqueous alteration product in the Martian meteorites. In addition, Lafayette's iddingsite may contain information about Martian groundwater in the very

**Table 4.** Chemical Model for Lafayette Iddingsite

	Iddingsite	Model for Iddingsite	Olivine	Mesostasis Glass
SiO <sub>2</sub>	49.1	52.4	32.4	76.8
TiO <sub>2</sub>	0.01	0.07	0.00	0.16
Al <sub>2</sub> O <sub>3</sub>	4.77	5.8	0.49	12.3
Cr <sub>2</sub> O <sub>3</sub>	0.00	0.0	0.01	—
FeO	29.01	28.1	51.0	0.04
NiO	0.10	0.07	0.16	—
MnO	0.48	0.52	0.94	—
MgO	13.90	8.4	14.65	0.72
CaO	0.70	0.4	0.18	0.56
Na <sub>2</sub> O	0.30	0.8	0.00	1.71
K <sub>2</sub> O	0.41	3.1	0.02	6.8
P <sub>2</sub> O <sub>5</sub>	0.09	0.4	0.05	0.8*
SO <sub>3</sub>	0.69	0.1	0.06	0.2*
Cl	0.36	0.05	0.02	0.1*

Iddingsite and olivine analyses from *Treiman et al.* [1993]. Glass analysis from *Berkley et al.* [1980], except stated otherwise. All normalized to 100% anhydrous. Model composition is 55% olivine + 45% mesostasis glass, the anhydrous equivalent of the 50% olivine + 40% mesostasis glass + 10% water model of the text.

\* From *Treiman et al.* [1993].

recent past; Lafayette itself crystallized from magma only 1300 m.y. ago, and its iddingsite formed less than 350 m.y. ago [Podosek, 1973; Swindle *et al.*, 1995]. Treiman *et al.* [1993] have already established a few physical and chemical constraints on the formation of Lafayette's iddingsite: it formed from liquid water; the temperature was  $<100^{\circ}\text{C}$ ; and the oxidation state was near the magnetite-hematite oxygen buffer. And, of course, they postulated that the composition approximates 50% olivine + 40% mesostasis glass + 10% water.

To arrive at any reasonable inferences about the composition of the parent water for the iddingsite, we need to understand how the iddingsite formed. If it crystallized from water, its composition should reflect element partitioning (and adsorption) between clay and water [Decarreau, 1985]. If the iddingsite grew rapidly, its composition should reflect kinetic factors. If the iddingsite includes salts from water trapped in the iddingsite, the analyzed compositions will reflect an unconstrained mixture of solids and dissolved salts.

### Iddingsite from Gel

Fortunately, the composition and structure of Lafayette's iddingsite are sufficiently distinctive to suggest a specific mechanism of formation. A hypothesis that seems to fit both its composition and structure is that the iddingsite formed originally as a silicate gel of nearly its present composition, which then crystallized (neofomed) to its present mineralogy.

First, the structure and mineralogy of Lafayette's iddingsite suggest that it was deposited from an aqueous medium [Treiman *et al.*, 1993]. The ferrihydrite in the iddingsite could reasonably form only at low temperatures ( $<100^{\circ}\text{C}$ ) under water-rich conditions; smectite itself is a hydrous silicate and the salt minerals in the iddingsite all suggest formation from water. In addition, Most of the iddingsite occurs as veins cutting olivine with boundaries along the olivines' (021) crystallographic planes; this is a common pattern in olivine dissolution [Treiman *et al.*, 1993 Figures 1a and 1b; DelVigne *et al.*, 1979].

However, the chemical composition of the iddingsite suggests that it did not crystallize directly from an aqueous fluid. The critical data here are that the iddingsite contains the transition metals Fe, Mn, Ni, Zn, and Co in effectively the same proportions as in the parent olivine (Tables 1 and 4) [Treiman *et al.*, 1993]. In smectite-water equilibria, however, the elements Ni, Zn, and Co are more strongly partitioned into smectite than is  $\text{Fe}^{2+}$ , and Mn is less strongly partitioned into smectite than is  $\text{Fe}^{2+}$  [Decarreau, 1985]. As none of these conditions are met by Lafayette's iddingsite and olivine, it seems unlikely that the iddingsite crystallized directly from waters that quantitatively dissolved the olivine. Similar, but smaller, fractionations should be present if the smectite grew at disequilibrium from solution [e.g., Lindstrom, 1983].

The iddingsite could reasonably have retained the element abundance ratios of its parent olivine if it formed by replacement of the olivine, but it did not. Most terrestrial iddingsite formed by replacement of olivine, and the iddingsite typically inherits much of the olivine's structural framework. For instance, the (001) octahedral layers of the smectites are commonly parallel to the (001) planes of the parent olivine [DelVigne *et al.*, 1979; Eggleton, 1984; Smith *et al.*, 1987; Banfield *et al.*, 1990]. However, Lafayette's iddingsite does not maintain any structural relationship with its host olivine and also occurs without an olivine host as intergranular films between plagioclase and pyroxene [Treiman *et al.*, 1993, Figures 1 and 2a].

Thus we arrive at an apparent contradiction: Lafayette's

iddingsite must have grown from aqueous solution to yield the observed minerals and textures; but the iddingsite could not have formed as crystalline silicates and oxides because it does not show the required elemental fractionations. A possible solution to this quandary is that the iddingsite formed originally as a silicate gel, which then crystallized to its present smectite-ferrihydrite mineralogy. Amorphous, gel-like materials can form during basalt weathering [Gooding, 1986; Eggleton, 1987; Gislason *et al.*, 1993]. And smectites crystallize readily from gels of their own compositions; gel crystallization is one of the standard laboratory methods for producing crystalline smectites [e.g., Klopprogge *et al.*, 1994]. We hypothesize that water, having entered Lafayette, dissolved both olivine and mesostasis glass nearly quantitatively. Salt minerals crystallized onto the walls of the dissolution cavities [e.g., Gooding *et al.*, 1991, Figure 5d]. The solution then gelled and crystallized in place to the smectite-ferrihydrite assemblage of the iddingsite. The extremely high  $\delta^{18}\text{O} = +14\text{‰}$  of the iddingsite [Romanek *et al.*, 1996] would have arisen from water - smectite fractionation as the gel crystallized.

### Fluid Composition

If Lafayette's iddingsite formed originally as a gel, then at least one constraint can be placed on the composition of the altering water: it was probably rich in  $\text{Mg}^{2+}$ . In the gel hypothesis, the iddingsite should contain whatever solutes were carried by the altering water, less what was lost as the gel crystallized. We estimate that the gel contained about 20% water by weight because the iddingsite now contains ~10% water and shows significant porosity that was probably a result of desiccation [Treiman *et al.*, 1993, Figures 2c, 2d, and 2e]. Thus, the composition of the original gel was approximately 45% olivine + 35% mesostasis glass + 20% water, and one can calculate solute concentrations in the water by mass balance.

The only element for which an actual concentration can be calculated is Mg because it is the only element which is more abundant in the real iddingsite than in the 50-40-10 model of iddingsite formation (Table 3). If the model is correct, ~8% MgO of the iddingsite's 14% MgO was contributed by the olivine, leaving ~6% of the iddingsite's MgO to be contributed from the solution. The solution could have contributed more Mg, but only this much was retained in the iddingsite. Thus the solution would have contained at least ~30 g MgO per 100 g water, a  $\text{Mg}^{2+}$  concentration of  $>0.7$  molar. Saturated solutions of  $\text{MgSO}_4$  or  $\text{MgCl}_2$  at  $100^{\circ}\text{C}$  contain about 0.7 molar  $\text{Mg}^{2+}$  [Weast, 1985]. Magnesium sulfate salts have been found in nakhlites [Gooding, 1992], and magnesium sulfate may be abundant in the Martian soil [Clark and Van Hart, 1981; Clark, 1993], so a Mg-rich solution seems reasonable.

### Conclusions

The INAA trace element analyses here have allowed a modest elaboration on the model that the average composition of Lafayette's iddingsite is essentially 50% olivine + 40% mesostasis glass + 10% water [Treiman *et al.*, 1993]. This model is consistent with abundances of Si, Al, Fe, Mn, Ni, Co, and Zn. The model's success, and the lack of element fractionations characteristic of smectite-water equilibria, are consistent with the original deposition of a silicate gel and its subsequent crystallization to the smectite and ferrihydrite of the iddingsite. Compared to the model's prediction, magnesium is significantly overabundant in the iddingsite (Table 3), and might

indicate that the altering waters were rich in  $Mg^{2+}$ . Mercury abundances are extremely high, probably as a result of terrestrial contamination.

It has been difficult to develop firm constraints on the composition of the waters involved in forming Lafayette's iddingsite. Little of the chemistry of the altering water can be disentangled from other chemical effects, including uncertainty in composition of the mesostasis glass, mineral admixtures in our iddingsite samples, the intrinsic variability of the iddingsite composition, and contamination on Earth. We have been able to develop no certain conclusions about the composition of the altering water; even the inference that it was rich in  $Mg^{2+}$  rests on a series of approximations and assumptions, untestable at this time. Even so, this failure to determine water composition does confirm the suggestion that iddingsite formation involved low water/rock ratios, not copious water flushing through Lafayette.

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