

Evidence for metasomatism of the lunar highlands and the origin of whitlockite

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Abstract—The presence of rare earth element-rich (REE-rich) whitlockite in the relatively primitive, REE-poor lunar highlands presents a paragenetic problem to the petrologist. Comparison with occurrences in mare basalt mesostasis assemblages demonstrates that whitlockite in the lunar highlands has not resulted from the crystallization of the residual interstitial melt. Furthermore, by the nature of REE partition coefficients for whitlockite, it is demonstrated that they cannot have crystallized from KREEPy melts. Rather, we suggest that the only lunar melt which contains high enough REE abundances is the basic immiscible melt derived from a KREEPy liquid. We envisage that whitlockite found in the lunar highlands is a post-cumulus metasomatic phase precipitated from a melt formed when urKREEP underwent silicate liquid immiscibility. The basic immiscible melt thus produced was able to percolate through the lunar highlands due to its low viscosity and act as a metasomatizing agent. Apatite is occasionally found as either epitactic overgrowths on the whitlockites, mantled by whitlockite, or in composite grains with whitlockite. In all cases, these apatites contain much lower REE contents than the corresponding whitlockite. Experimental results suggest either (1) apatite and whitlockite have essentially the same REE Kd's, or (2) apatite has lower REE Kd's than whitlockite. Both scenarios can be used to explain certain apatite-whitlockite associations. For example, apatite and whitlockite having the same REE Kd's can explain the situation where apatite appears to have crystallized after whitlockite. This is because apatite crystallization requires a certain level of halogens in the melt. These levels are only realized after whitlockite crystallization, which enriches the residual melt in F and Cl, but depletes it in the REEs. Therefore, when apatite crystallizes it will contain lower REE abundances than whitlockite. However, relationships where apatite and whitlockite appear to have co-crystallized, or apatite crystallized before whitlockite, require apatite to have lower REE Kd's. Even if both experimental results are correct, the whitlockite parental melt still requires REE abundances greater than in KREEP/urKREEP. The parental melt has similar REE abundances to a calculated REEPy immiscible melt from KREEP/urKREEP.

INTRODUCTION

THE MOON IS A RELATIVELY basic, primitive planetary body, but contains highly evolved minerals and lithologies which present fundamental problems to the petrologist. Evolved lithologies and minerals include urKREEP (e.g., MEYER and HUBBARD, 1970; MEYER et al., 1971; WARREN and WASSON, 1979), granite (e.g., BLANCHARD et al., 1977; WARREN et al., 1983a, 1987), quartz-monzodiorite (QMD; e.g., RYDER, 1976; MARVIN et al., 1990), and the presence of REE-rich whitlockite in the primitive lunar highlands (e.g., LINDSTROM et al., 1984, 1985; SHERVAIS et al., 1984). UrKREEP is considered to be the final (last 1%) dregs of the Lunar Magma Ocean (WARREN and WASSON, 1979; WARREN, 1985), and granite is the immiscible product of a KREEPy magma (HESS and RUTHERFORD, 1974; HESS et al., 1975; NEAL and TAYLOR, 1988, 1989). Quartz-monzodiorite contains greater abundances of the REE than the high-K KREEP defined by WARREN (1988) and appears not to be an immiscible product from such a composition, but an extreme fractionate (TAYLOR et al., 1980). NEAL and TAYLOR (1989) proposed that the highly evolved lunar phosphates crystallized from the REEP-Fraction after KREEP experiences silicate liquid immiscibility (SLI), and lunar granite represents the K-Fraction.

NEAL and TAYLOR (1988, 1989) suggested that viscosity differences between the K- and REEP-Fractions, coupled with post-SLI crystal fractionation of ilmenite and fayalite, allows

the REEP-Fraction to percolate upward, metasomatizing the lunar highlands. These authors concluded that such a process could account for the presence of highly evolved phosphates in the primitive lunar highlands. LINDSTROM et al. (1984, 1985) and SHERVAIS et al. (1984) suggested that these whitlockites were the products of variable degrees of KREEP assimilation by the parental magma, whereas NEAL et al. (1987), SHERVAIS (1990), and SHERVAIS and VETTER (1991) concluded that the intercumulus liquid of these highland plutons *auto-metasomatized* the plagioclase-rich cumulates. However, HESS et al. (1990) suggested that the presence of whitlockite in the lunar highlands is due to extreme fractional crystallization of the inter-cumulus liquid, without SLI. WARREN et al. (1983b) and LINDSTROM et al. (1984) suggested that there was evidence for the formation of whitlockite in the highland lithologies from both intercumulus liquid crystallization and metasomatism. Both WARREN et al. (1983b) and LINDSTROM et al. (1984) suggested that if metasomatism produced the highland whitlockites, then the metasomatizing agent was either KREEP or urKREEP, even though the incompatible elements are not present in KREEPy proportions (WARREN et al., 1983b). Whatever the exact details, all authors are agreed that the whitlockite has crystallized from an evolved silicate melt. This was demonstrated by HESS et al. (1990) in experiments to determine the FeO/MgO distribution coefficient between whitlockite and liquid. Their results indicate that a whitlockite with an MG# of 0.65

will coexist with a liquid of MG# 0.26 at 1050°C and 0.12 at 1000°C.

The purpose of this paper is to investigate the origin(s) of this minor yet significant mineral, whitlockite. We will use available experimental results of elemental partitioning between whitlockite and melt and between two immiscible liquids to suggest a metasomatic alternative for the origin of whitlockite.

THE OCCURRENCE OF WHITLOCKITE

REE-rich whitlockite has been described from alkali anorthosites (WARREN et al., 1983b; SHERVAIS et al., 1984; GOODRICH et al., 1986; NEAL et al., 1990), ferroan anorthosites (e.g., GOODRICH et al., 1985), Mg-suite anorthosites (LINDSTROM et al., 1984; NEAL et al., 1990), the alkali gabbro suite from Apollo 16 (e.g., LINDSTROM et al., 1984; JAMES et al., 1987), and the Apollo 15 quartz-monzodiorite (e.g., MARVIN et al., 1990; LINDSTROM et al., 1991), as well as in the mesostases of mare basalts (e.g., ALBEE and CHODOS, 1970; BROWN et al., 1971). It has a variety of textural occurrences: (1) as an interstitial phase (LINDSTROM et al., 1984); (2) as an inclusion in plagioclase (e.g., SHERVAIS et al., 1984; JAMES et al., 1987); (3) as an inclusion in apatite, possibly a reaction relationship (GOODRICH et al., 1986); and (4) forming composite grains with apatite (e.g., JAMES et al., 1987). These must be taken into account in any model of whitlockite crystallization.

The presence of apparent phosphate inclusions in the constituent minerals of the various highland suites has been explained by SHERVAIS and VETTER (1991) in the following way. Post-cumulus fracturing of the cumulus minerals occurred, and the phosphate parental magma was injected along these lines of weakness. This may have been auto-metasomatism by the post-cumulus melt (NEAL et al., 1987; SHERVAIS and VETTER, 1991) or metasomatism by an unrelated melt (NEAL and TAYLOR, 1989). After phosphate crystallization, annealing occurred producing the apparent inclusion, without having whitlockite crystallizing first. Furthermore, both the plagioclase and phosphate may have originated from the metasomatic fluid (NEAL and TAYLOR, 1989; LU et al., 1989).

WHITLOCKITE COMPOSITIONS

We have compiled a set of whitlockite compositions from highlands lithologies at the Apollo 14, 16, and 17 sites, as well as from lunar meteorite ALHA81005 and mare basalts from the Apollo 12 and 14 sites (Table 1). Where partial REE analyses have been reported, we have estimated certain REE abundances (where possible) by extrapolation of the REE profile. These extrapolated values are denoted by parentheses in Table 1. The whitlockites reported in Table 1 are from ferroan (GOODRICH et al., 1985), alkali (WARREN et al., 1983b; SHERVAIS et al., 1984; GOODRICH et al., 1986), and Mg anorthosites (LINDSTROM et al., 1984; NEAL et al., 1987, 1990), as well as alkali gabbroites (JAMES et al., 1987), and the mesostases of mare basalts (ALBEE and CHODOS, 1970; BROWN et al., 1971; FRONDEL, 1975; and references therein).

The REE compositions of whitlockite from these various rock types generally overlap (Fig. 1) but exhibit a range in REE abundances within each grouping. All LREE are present between 9500 and 36000 times chondritic abundances, whereas the HREE are between 3000 and 20000 times chondritic levels. With such high REE abundances, analysis of whitlockites can be conducted on the electron microprobe. LINDSTROM et al. (1985) reported three ion microprobe (IMP) analyses of whitlockites (two from Apollo 14 anorthosites—one magnesian and one alkali, and one from an Apollo 16 alkali gabbroite), which agree well with the EMP (Electron MicroProbe) analyses reported by SHERVAIS et al. (1984) and LINDSTROM et al. (1984). These IMP analyses confirm that whitlockite possesses a large negative Eu anomaly. All are LREE-enriched (La_N/Yb_N and $La_N/Sm_N > 1$). Because of the low Eu abundances in whitlockite, it was not accurately detected by the EMP and as such, Eu is not reported for whitlockites analyzed by EMP as it is generally below detection level. However, we infer from IMP analyses that all whitlockite REE patterns contain a negative Eu anomaly but have not shown this by extrapolation on subsequent REE plots.

CRYSTALLIZATION OF WHITLOCKITE

The MgO and FeO contents reported in Fig. 2 demonstrate a range in MG# for the whitlockite compositions (Table 1). For example, all whitlockites generally have MG#'s > 80 , with only those found in alkali anorthosites and basaltic mesostases having MG#'s < 80 . These compositional differences may be due to different crystallization temperatures which require different partition coefficients for FeO and MgO partitioning between whitlockite and melt (HESS et al., 1990). However, these variable whitlockite MG#'s may also be a reflection of different bulk compositions of the individual parental melts, or equilibration with the host rock. Likewise, the variable REE abundances in the whitlockites from these various lithologies (Fig. 1) may be due to similar considerations (LINDSTROM et al., 1984). The whitlockites from basaltic mesostases (e.g., ALBEE and CHODOS, 1970; BROWN et al., 1971; FRONDEL, 1975, and references therein) practically span the range defined by whitlockites from the highland lithologies (Fig. 1). With such high MG#'s, it may be difficult to envisage whitlockite crystallization from an evolved melt. However, HESS et al. (1990) suggested that a whitlockite of relatively high MG# could crystallize from an evolved melt.

GOODRICH et al. (1986) noted that the presence of REE-rich phosphates in all major pristine highlands suites suggested that the REEs and P can be decoupled from other elements that would normally show similar chemical behavior. However, GOODRICH et al. (1986) concluded that the differences between whitlockite REE profiles are not as large as would be expected from the differences in major-element chemistry of the parental magmas which formed the various highlands suites. These authors further suggested that whitlockites in the highland lithologies could be a metasomatic product produced by a basic immiscible melt. Such a metasomatizing agent could account for the dichotomy between the REEs and P and other similar elements.

Table 1: Whitlockite Compositions. Where partial analyses have been reported, the REE abundances have been estimated by extrapolation of the REE profile. Extrapolated values in ().

N.o.	73216	73216	14321*	73216	14304	14305*	14313	14321	67975*	67975*	88105	88105	12040	12003	12013.10	14310.20B	12040	12036	
Ref.	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	
Rock	M	M	M	M	A	A	A	A	A	A	A	A	B	B	B	B	B	B	
SiO ₂	0.92	0.78	1.16	1.10	1.81	1.81	2.61	1.51	0.99	0.95	1.13	0.68	0.73	2.30	0.77	1.38	3.35	2.30	2.13
FeO	3.01	3.23	2.97	3.20	2.98	3.12	2.54	2.78	3.22	3.01	2.85	3.29	3.22	3.32	3.43	2.45	1.88	3.32	3.80
MgO	40.7	42.9	40.1	42.3	40.9	40.8	42.5	44.7	38.5	39.1	38.0	42.8	43.2	42.3	38.6	38.7	42.3	42.3	42.3
CaO	43.5	44.1	44.1	43.5	44.3	43.3	42.6	44.0	42.1	42.6	44.6	44.5	44.3	44.8	44.5	43.2	44.3	44.3	42.5
P ₂ O ₅	1.36	0.74	0.66	0.77	0.75	0.79	0.68	0.73	0.92	0.66	1.16	0.62	0.58	0.38	0.64	1.09	1.02	0.38	0.50
Li ₂ O	3.73	2.11	1.76	2.26	1.87	2.86	(1.42)	(1.28)	2.43	2.40	3.17	(1.14)	(1.05)	1.45	2.20	(2.25)	2.86	1.38	1.69
CaO ₂	(0.33)	(0.21)	0.22	(0.24)	0.22	(0.26)	(0.18)	(0.17)	0.30	(0.26)	(0.30)	(0.16)	(0.14)	0.44	0.54	0.76	0.74	0.44	0.30
P ₂ O ₅	(1.52)	(1.08)	1.07	(1.22)	1.00	1.55	1.34	1.29	1.11	1.47	1.89	1.95	1.10	1.37	1.51	2.15	2.05	1.37	(1.04)
Nd ₂ O ₃	0.56	0.43	0.29	0.51	0.24	0.48	0.36	0.35	0.32	0.40	0.66	0.57	0.34	0.35	0.30	0.66	1.00	0.30	(0.26)
Sm ₂ O ₃	0.69	0.51	0.26	0.56	0.22	(0.49)	(0.33)	(0.26)	0.32	(0.56)	(0.48)	(0.32)	(0.26)	0.60	0.67	1.11	0.84	0.60	0.50
Gd ₂ O ₃	0.37	0.37	0.34	0.34	0.26	0.54	(0.33)	(0.28)	0.46	0.80	0.70	(0.37)	(0.33)	0.51	0.43	0.38	(0.64)	0.51	0.37
Dy ₂ O ₃	(0.17)	(0.17)	0.19	(0.12)	0.14	0.30	0.24	0.23	0.21	0.26	(0.54)	(0.28)	0.30	0.25	0.20	0.15	(0.17)	(0.50)	(0.14)
Er ₂ O ₃	0.20	0.20	0.17	0.11	0.11	0.25	0.16	0.13	0.27	(0.55)	(0.24)	0.13	0.10	(0.10)	(0.16)	(0.30)	(0.13)	(0.07)	(0.07)
Yb ₂ O ₃	1.95	1.85	2.64	1.96	1.83	—	2.84	2.35	3.86	5.00	2.86	2.61	2.17	1.93	3.31	3.01	2.17	2.17	2.28
Y ₂ O ₃	99.01	98.68	96.31	98.19	96.85	96.45	97.82	98.30	100.43	95.81	99.23	98.17	99.29	100.02	99.51	100.50	100.16	97.78	97.78
TOTAL	8.93	5.82	4.96	6.13	4.80	7.88	5.15	4.72	4.51	6.83	8.32	8.85	8.85	5.35	6.88	9.07	5.31	4.87	4.87
SUM REE	88.1	82.0	83.8	78.0	83.5	71.4	64.1	76.6	85.3	85.3	89.6	88.7	72.0	88.8	76.0	50.0	72.0	76.1	76.1
MG#	6310	7702	6566	8700	8906	6736	5798	6225	10760	5628	9891	5287	4946	(3240)	9294	8697	(3240)	4263	4263
La	11596	18014	20624	19295	21912	24418	(10672)	(10928)	28430	20490	27064	(9733)	(8965)	12380	18783	24418	1782	14429	14429
Ce	(2820)	(1794)	2566	(2051)	2609	(2222)	(1538)	(1453)	3515	(2222)	(2563)	(1367)	(1196)	3760	4614	6494	6323	3760	2563
Pr	(13032)	(9259)	12475	(10460)	11662	13289	11488	11660	17108	16204	16718	9431	8659	11746	12946	18433	17146	(8916)	(8916)
Nd	4829	3708	3315	4398	2827	4139	3104	3018	4608	5691	4915	2932	2587	5691	8623	8796	2587	(2242)	(2242)
Sm	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
Eu	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
Gd	5986	4425	3026	4838	2500	4251	(2863)	(2429)	3661	(4858)	(4164)	(2776)	(2256)	5206	5813	9630	5206	4338	4338
Dy	3224	3224	3890	2962	3030	4705	(2875)	(2440)	5329	6099	(3224)	(2875)	4444	3747	3311	(5576)	4444	3224	3224
Er	(1487)	(1487)	2173	(1049)	1624	2624	2099	2011	3004	(4722)	(2449)	2624	2186	1749	1312	(1992)	(1749)	(1749)	(1749)
Yb	1756	1756	1903	966	1252	2195	1405	1317	3070	(4830)	(2108)	1142	878	(878)	(1012)	(2635)	(1142)	(645)	(645)
Y	15355	14568	20788	15434	14410	23623	35435	11812	30395	39372	25521	20552	9449	7874	26064	23702	17087	17087	17814

A = Alkali Anorthositic; B = Basalt; F = Ferrugin Anorthositic; G = Alkali Gabbroitic; M = Mg Anorthositic.

References: 1 = NEAL *et al.* (1990); 2 = LINDSTROM *et al.* (1985); 3 = LINDSTROM *et al.* (1984); 4 = SHERVALIS *et al.* (1984); 5 = JAMES *et al.* (1987); 6 = GOODRICH *et al.* (1985); 7 = GOODRICH *et al.* (1986); 8 = WARREN *et al.* (1983b); 9 = BROWN *et al.* (1971); 10 = PRONDEL (1975); 11 = KEIL *et al.* (1971).

* = Ion microprobe analysis.

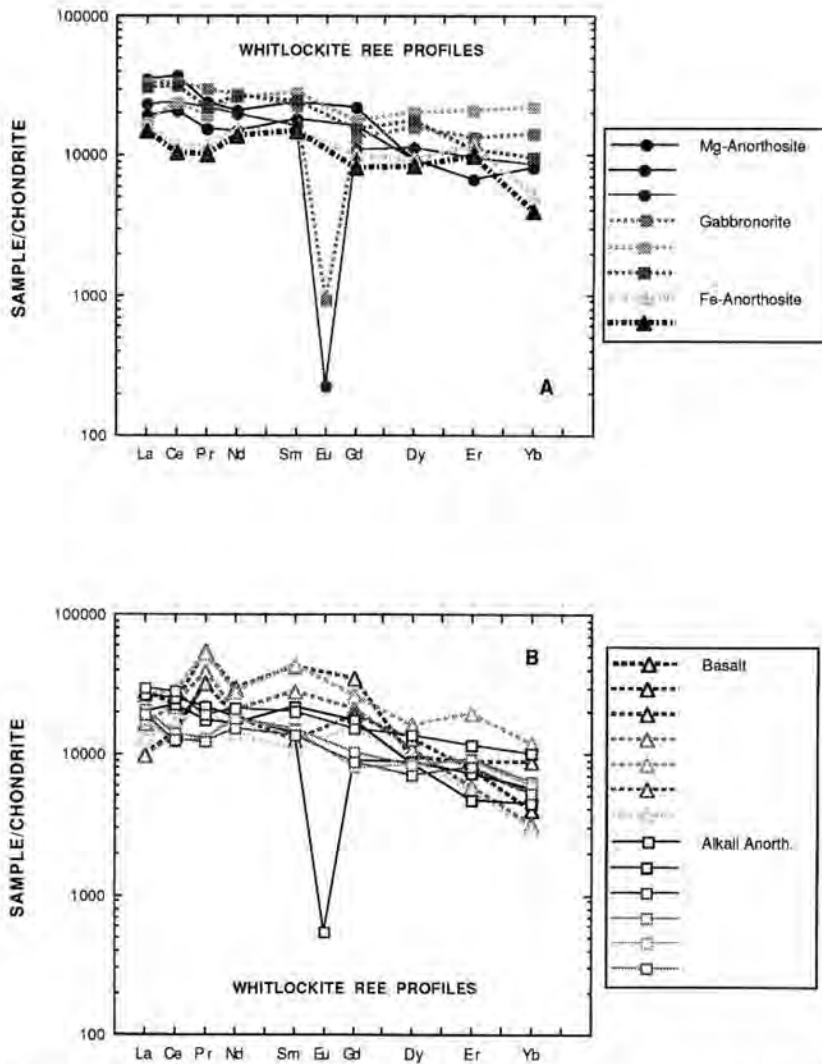


FIG. 1. Lunar whitlockite REE. Europium only plotted for ion microprobe analyses (LINDSTROM *et al.*, 1985). Data from: NEAL *et al.* (1990); LINDSTROM *et al.* (1985); LINDSTROM *et al.* (1984); SHERVAIS *et al.* (1984); JAMES *et al.* (1987); GOODRICH *et al.* (1985, 1986); WARREN *et al.* (1983b); BROWN *et al.* (1971); KEIL *et al.* (1971); FRONDEL (1975), and references therein.

NEAL and TAYLOR (1988, 1989) have expanded this suggestion of GOODRICH *et al.* (1986) with their K-Fraction (granite) and REEP-Fraction hypothesis, which can be achieved by urKREEP experiencing immiscibility. NEAL and TAYLOR (1990) have concluded, using the partition coefficients of DICKINSON and HESS (1983), that whitlockites in the lunar highlands crystallized from a REE-rich melt akin to the basic immiscible melt produced by the immiscibility of urKREEP. However, the relevance of these whitlockite/liquid partition coefficients has recently been questioned (HESS *et al.*, 1990; HESS, *pers. comm.*, 1990).

WHITLOCKITE/LIQUID REE PARTITION COEFFICIENTS

In order to evaluate the REE composition of the melt from which whitlockite crystallized, the REE partition coefficients

(Kd's) must be accurately known. DICKINSON and HESS (1983) reported whitlockite partition coefficients for Ce (9.5), Sm (9.5), Eu (2.6), Gd (11.4), and Yb (5.8), and MCKAY *et al.* (1987) reported those for Nd and Sm. Recently, it has been suggested that the whitlockite REE Kd's may be as high as 40 (HESS, *pers. comm.*, 1990). GOODRICH *et al.* (1986) questioned the validity of regarding the REEs in whitlockite as being trace elements, suggesting that they should be considered as major elements. However, although the REEs in whitlockite are present in the wt% level, the experimental results of MCKAY *et al.* (1987) indicated that there was no unexpected deviation from Henry's Law behavior, and that experimental whitlockite REE partition coefficients may be safely extrapolated to natural concentrations.

The results of MCKAY *et al.* (1987) demonstrated that the partition coefficient for a particular REE in whitlockite is

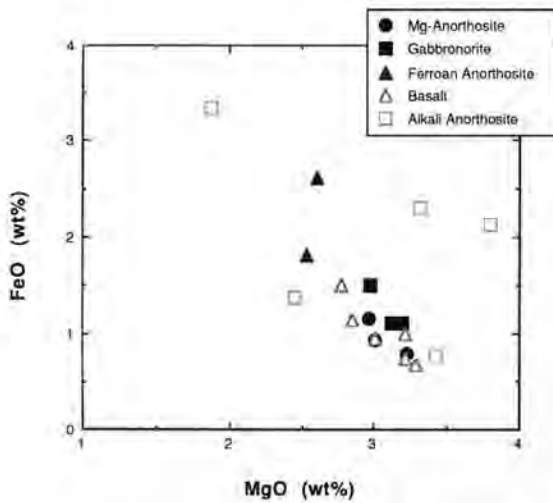


FIG. 2. MgO vs. FeO for lunar whitlockites. See text for discussion. Data sources as in Fig. 1.

dependent upon total REE concentration. A similar effect has previously been observed for garnet (HARRISON, 1981). Also, this study confirmed that the Kd for Nd and Sm was approximately the same, as suggested by DICKINSON and HESS (1983) who concluded that there are no appreciable differences in Kd's for the LREEs. To obtain whitlockite Kd's for the compositions presented in Table 1, we have used the rationale of MCKAY et al. (1987) to anchor the Nd partition coefficient. In Fig. 3, the variation of Nd Kd with total REE concentration in whitlockite is presented (Fig. 6 of MCKAY et al., 1987). We have determined the Nd Kd for each of the whitlockites presented in Table 1. The total REE concentration for each whitlockite (Table 1) was derived by extrapolation of the REE profile produced from direct analysis of a number of the REEs. The Nd Kd was taken from the middle

of the field shown in Figure 3. We assumed that the REE profile for whitlockite/liquid Kd's presented by DICKINSON and HESS (1983) was correct (i.e., the differences in Kd between the various REEs were consistent) and that there was no difference in Kd's for La, Ce, Nd, and Sm. In this way, we have interpolated whitlockite/liquid REE Kd values (Fig. 4 and Table 2) for each of the whitlockite compositions used in this study (Table 1).

Whitlockite Equilibrium Liquids

The composition of the whitlockite equilibrium liquid depends upon the composition of the parental basalt (HESS et al., 1975; RUTHERFORD et al., 1976). If the original magma was a mare basalt, then a basic immiscible melt will be ferrobasaltic in composition. Some KREEPy compositions will yield a basic immiscible melt composition equivalent to quartz monzodiorite. For our model, we will assume that the Kd's are appropriate for KREEPy basalts. Using the whitlockite/liquid Kd's for the REEs as defined previously (Table 2), the composition of the melt with which the whitlockite was last in equilibrium can be estimated (Fig. 5 and Table 3). The La abundances in these equilibrium liquids range from approximately 600 to 3000 times chondrite levels. Equilibrium liquids for whitlockites in the ferroan anorthosites generally contain the lowest REE abundances (Fig. 5). Eu abundances have only been accurately determined for 3 whitlockites analyzed by the ion microprobe (LINDSTROM et al., 1985) and demonstrated that the whitlockites (Fig. 1) and the corresponding equilibrium liquids (Fig. 5) possess large negative Eu anomalies. Because of the lack of accurate Eu data for whitlockite analyzed by EMP, we have hesitated in extrapolating these plots to include Eu for all whitlockites in Table 1. Generally, all equilibrium liquids exhibit LREE-enriched profiles, with negative correlations for both the LREEs and HREEs, although some exhibit relatively flat HREE profiles (Fig. 5).

The origin of the whitlockite equilibrium liquids can be

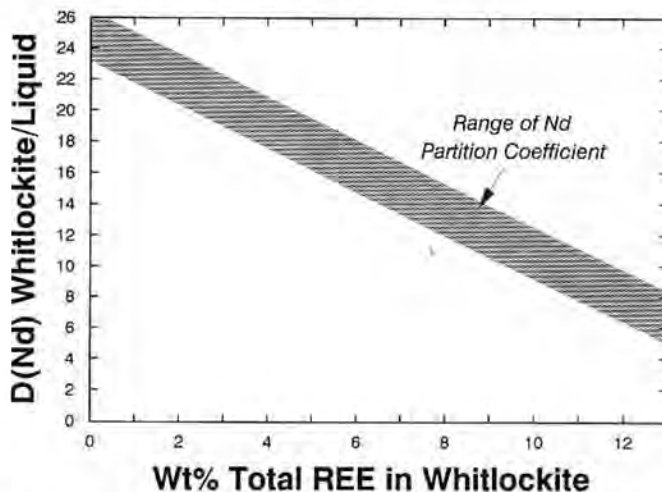


FIG. 3. Range of Nd partition coefficient for whitlockite as a function of total REE contents. Data from MCKAY et al. (1987).

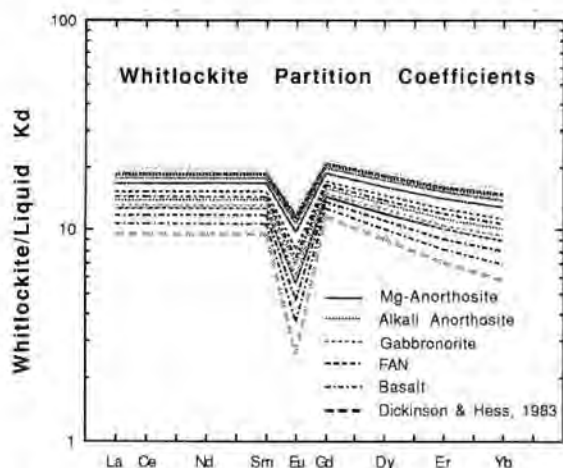


FIG. 4. REE/liquid partition coefficients for each reported lunar whitlockite. The patterns have been constructed using the data of DICKINSON and HESS (1983) and absolute values after the rationale of MCKAY et al. (1987).

examined by normalizing the REE abundances to certain KREEP (or urKREEP) compositions. These KREEPy compositions are from (1) MORRIS et al. (1990): analysis of 14001,28.3, a granite fragment intruded by a basic immiscible melt, possibly the first pristine urKREEP sample identified; (2) NEAL and TAYLOR (1989): the average calculated urKREEP composition from the K-Fraction/REEP-Fraction hypothesis; and (3) WARREN (1988): a compilation of a high-K KREEP composition from lunar samples. These KREEP compositions are presented in Table 4, and results of the normalization are presented in Fig. 6 a-f.

As can be seen from Fig. 6, the whitlockite equilibrium liquids are all higher than the measured or calculated KREEP and urKREEP compositions. It is evident that both the measured KREEP compositions of MORRIS et al. (1990) (Fig. 6 a,b), the average calculated urKREEP of NEAL and TAYLOR (1989) (Fig. 6 c,d), and the high-K KREEP of WARREN (1988) (Fig. 6 e,f) cannot be parental to the lunar whitlockite compositions, because when normalized to these KREEP/urKREEP compositions, practically all equilibrium liquids yield values > 1 .

PETROGENESIS OF LUNAR WHITLOCKITE

It is evident that whitlockite present in the lunar highlands and in basaltic mesostases crystallized from a highly REE-enriched melt. The question remains, How is the REE-rich melt formed? The presence of whitlockite in the basaltic mesostases would appear to suggest that extreme fractional crystallization of a basaltic magma is responsible. In these basalts, whitlockite and/or apatite is commonly found in association with ilmenite, fayalite, silica (cristobalite), and K-rich glass (TAYLOR et al., 1971) and whitlockite contains higher REE abundances than apatite (e.g., ALBEE and CHODOS, 1970; BROWN et al., 1971). This assemblage is consistent with immiscibility of the residual basaltic magma. While both basic and acidic glasses are not normally present, the assemblage fayalite-ilmenite-phosphate-cristobalite is consistent with the

crystallization of a basic glass resulting from immiscibility (e.g., LU et al., 1989). TAYLOR et al. (1971) reported the presence of such a mesostasis assemblage in basalt 12063,9 and concluded that the modal proportions of fayalite and cristobalite represented the crystallized equivalents of a ferroproxenitic or basic immiscible melt. Viscosity differences between acidic and basic immiscible melts ($\sim 30,000$ vs. 11 poise, respectively; NEAL and TAYLOR, 1989) suggest that crystallization of the basic immiscible melt will be more readily achieved than that of the acidic equivalent. Indeed, TAYLOR et al. (1971) concluded that the K-rich glass inclusions observed in the outer margins of mesostasis fayalite represented globules of immiscible granitic liquid originally accumulated around the margins of pools of the more abundant ferroproxenitic or basic melt. The relatively fast cooling of the lunar basalts at the surface means we only see immiscibility and crystallization/quenching of the immiscible glasses in a microcosm. At depth, the cooling would be slower, allowing for more extensive development of immiscible melts. However, if whitlockite in the lunar highlands formed from the *in situ* crystallization of intercumulus liquid, similar mineral assemblages to those in the basaltic mesostases would be expected. This is not the case, as whitlockite is commonly found in isolation (e.g., LINDSTROM et al., 1984; SHERVAIS et al., 1984; NEAL et al., 1987), or associated with only apatite, ilmenite, or pyroxene (e.g., JAMES et al., 1987; GOODRICH et al., 1986).

From the close examination of basaltic mesostases, it is evident that the process of silicate liquid immiscibility may be fundamental to the crystallization of whitlockite. This process preferentially partitions Si, Al, K, Rb, Cs, and Na into the acidic immiscible melt, whereas Ti, Cr, Fe, Mn, REE, P, Ca, Th, U, Zr, F, and Cl are preferentially partitioned into the basic immiscible melt (HESS and RUTHERFORD, 1974; HESS et al., 1975; WATSON, 1976). Note that the halogens, Ca, REEs, and P are preferentially partitioned into the basic immiscible melt. As stated above, in these basaltic mesostases are the crystallized equivalents of a basic immiscible melt or REEP-Fraction (NEAL and TAYLOR, 1988, 1989) and a glassy granitic melt or K-Fraction. In order to test the validity of this observation, the REE abundances of the basic immiscible melts have been calculated from the three KREEP compositions reported in Table 4. The basic immiscible melt compositions (Table 4) were estimated using the liquid-liquid K_d 's ($D_{b/a}$) of RYERSON and HESS (1980) for both low- and high- P_2O_5 systems. However, determination of elemental abundances requires a knowledge of the granite-ferrobalt immiscible liquids ratio, which is difficult to quantify as this ratio changes with temperature (RUTHERFORD et al., 1976). For simplicity, we will assume that the ferrobalt is dominant in that the bulk composition is just inside the immiscibility gap. This is because the whitlockite is unlikely to be from the granite due to viscosity considerations. Therefore, by simply multiplying the elemental abundance in the KREEP composition by the two-liquid K_d ($D_{b/a}$), the elemental abundance in the basic immiscible melt can be estimated. Although only two-liquid K_d 's were reported for La and Yb by RYERSON and HESS (1980), we estimated those for the other REEs by extrapolation (Table 4). This is a feasible approach because the REEs exhibit little fractionation between each other (ELLISON and HESS, 1989). However, the

Table 2: Estimated whitlockite/melt partition coefficients.

No.	73216 ,36	73216 ,42	14321* ,1211	73216 ,38	14305* ,400	14304 ,86	14305 ,303	14313 ,70	14321 ,1060
Rock	M	M	M	A	A	A	A	A	A
La	12.5	16.6	17.6	16.6	18.0	13.8	11.5	18.2	18.6
Ce	12.5	16.6	17.6	16.6	18.0	13.8	11.5	18.2	18.6
Nd	12.5	16.6	17.6	16.6	18.0	13.8	11.5	18.2	18.6
Sm	12.5	16.6	17.6	16.6	18.0	13.8	11.5	18.2	18.6
Eu	5.6	9.7	10.7	9.7	11.1	6.9	4.6	11.3	11.7
Gd	14.4	18.5	19.5	18.5	19.9	15.7	13.4	20.1	20.5
Dy	12.0	16.1	17.1	16.1	17.5	13.3	11.0	17.7	18.1
Er	10.0	14.1	15.1	14.1	15.5	11.3	9.0	15.7	16.1
Yb	8.8	12.9	13.9	12.9	14.3	10.1	7.8	14.5	14.9

A = Alkali Anorthosite; B = Basalt; F = Ferroan Anorthosite; G = Alkali Gabbroanorthite; M = Mg Anorthosite.

No.	67975 ,136	67975 ,44Nm	67975 ,44Nf	88105 ,32	88105 ,32	12040	12003	12013,10	14310,20B
Rock	G	G	G	F	F	B	B	B	B
La	15.0	13.0	12.6	18.6	19.4	16.6	14.3	11.6	10.6
Ce	15.0	13.0	12.6	18.6	19.4	16.6	14.3	11.6	10.6
Nd	15.0	13.0	12.6	18.6	19.4	16.6	14.3	11.6	10.6
Sm	15.0	13.0	12.6	18.6	19.4	16.6	14.3	11.6	10.6
Eu	8.1	6.1	5.7	11.7	12.5	9.7	7.4	4.7	3.7
Gd	16.9	14.9	14.5	20.5	21.3	18.5	16.2	13.5	12.5
Dy	14.5	12.5	12.1	18.1	18.9	16.1	13.8	11.1	10.1
Er	12.5	10.5	10.1	16.1	16.9	14.1	11.8	9.1	8.1
Yb	11.3	9.3	8.9	14.9	15.7	12.9	10.6	7.9	6.9

No.	12040 ,39	12036 ,9	Dickinson & Hess, 1983
Rock	B	B	
La	16.6	17.6	9.5
Ce	16.6	17.6	9.5
Nd	16.6	17.6	9.5
Sm	16.6	17.6	9.5
Eu	9.7	10.7	2.6
Gd	18.5	19.5	11.4
Dy	16.1	17.1	9.0
Er	14.1	15.1	7.0
Yb	12.9	13.9	5.8

magnitude of the two-liquid REE Kd's is strongly dependent upon temperature. ELLISON and HESS (1989) demonstrated a change in La $D_{b/a}$ from 3.5 at 1450°C to 10.8 at 1350°C. Therefore, the REE concentrations of the basic immiscible melts may be higher than we state here, but for our modelling purposes, it will not affect the results. Rather, such an observation will actually support our model for whitlockite paragenesis.

The whitlockite equilibrium liquids have been normalized to the basic immiscible melts calculated from the KREEP compositions using both low- and high-P₂O₅ system liquid/liquid Kd's in Table 4 (Fig. 7 a-f). If such immiscible melts are likely parents for whitlockite, the resulting profiles should be close to unity. Whitlockite equilibrium liquid REE abundances show remarkable similarity to those of calculated basic immiscible melts from urKREEP compositions (NEAL and TAYLOR, 1989; MORRIS et al., 1990) using experimental liquid/liquid Kd's from a low-P₂O₅ system (i.e., normalized values are between 0.25 and 1.5 - Fig. 7 a-d). When the high-K KREEP composition of WARREN (1988) and the low-P₂O₅ liquid-liquid Kd's are used, ratios are generally >1 (Fig. 7 e, f), although equilibrium liquids of the whitlockites from the Ferroan Anorthosites fall below 1. Those basic immiscible melt compositions calculated from the three KREEP/urKREEP compositions using liquid/liquid Kd's from a high-P₂O₅ system are too enriched in the REEs and normalized values are <1 (Fig. 7). Although model dependent, this sug-

gests that the parental magma did not precipitate whitlockite until after immiscibility. However, MURRELL et al. (1984) reported whitlockite REE Kd's which are less than half those reported in Table 2, but these were determined at temperatures of 1200°C and for liquids of diopside-plagioclase composition. Therefore, absolute values reported by MURRELL et al. (1984) are of no use in a ferrobasaltic system at 900-1000°C, only the shape of the Kd profile is important (ELLISON and HESS, 1989). It is evident that further experimental work into phosphate/liquid REE partitioning is required.

GEOLOGICAL SETTING

The modelling outlined previously is dependent upon a number of unconstrained parameters (e.g., relevant liquid/liquid Kd's, temperature, granite-ferrobasalt ratio, etc.), but illustrates that whitlockite crystallized from a REE-rich, evolved melt. We recognize that immiscibility will concentrate P₂O₅ into our REEP-Fraction, increasing normative whitlockite and this has the ability to lower whitlockite/liquid REE Kd's; but if, as we assume, the whitlockite parental liquid is close to the pre-SLI composition (i.e., just inside the immiscibility field), the whitlockite REE Kd's will not be radically affected (HESS, pers. comm., 1991). It is the goal of this paper to outline a model for whitlockite crystallization in the lunar environment within the confines of present experimental data and current unconstrained parameters.

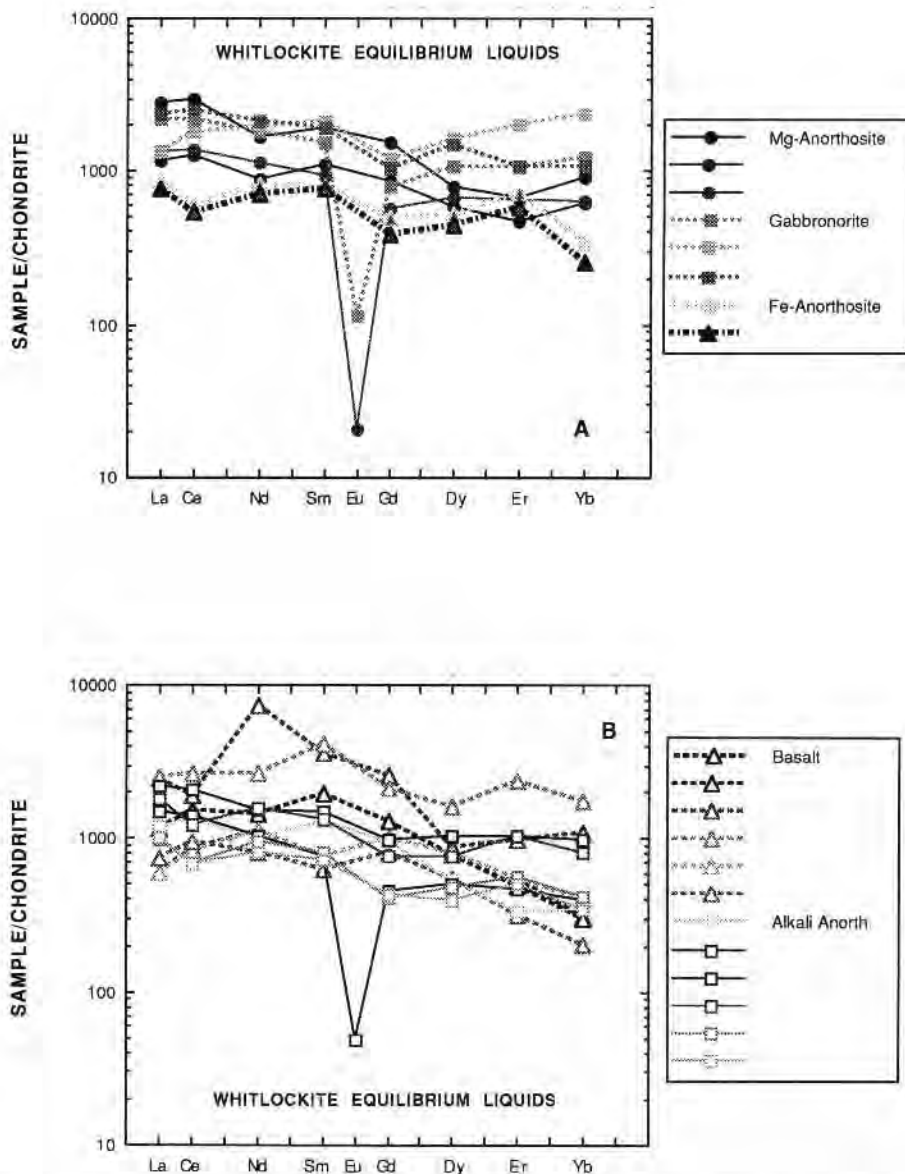


FIG. 5. REE patterns of the equilibrium liquids (i.e., the liquid with which each whitlockite reported here was last in equilibrium with) for lunar whitlockites. These have been calculated from the analyzed REE compositions and calculated partition coefficients.

Within this framework, our illustrative model demonstrates the similarity of whitlockite equilibrium liquid REE profiles to those of basic immiscible melts from several probable KREEP/urKREEP compositions. This suggests that SLI was an important process for whitlockite crystallization and that whitlockite may not have crystallized until after immiscibility.

On the basis of whitlockite equilibrium liquids, we have argued that the presence of whitlockite in the lunar highlands cannot be solely due to the crystallization of intercumulus liquid. Even so, the lack of the required mesostasis mineral assemblage in the lunar highlands demonstrates that the *in situ* intercumulus liquid did not undergo SLI to produce the parental liquid for whitlockite crystallization. On the basis of the calculated whitlockite/liquid K_d 's, a basic immiscible melt derived from KREEP/urKREEP by the process of SLI

is the only liquid which contains high enough REE abundances, on the basis of the whitlockite/liquid K_d 's presented here, to account for observed whitlockite compositions. In the case of basaltic mesostases, the incompatible element abundances in the residual magma must probably approach KREEPy levels prior to immiscibility in order to generate the observed whitlockite REE abundances. Are we seeing the lunar magma ocean in microcosm?

A metasomatic model is proposed in order to account for the presence of whitlockite in the lunar highlands. Observations of metasomatized terrestrial rocks demonstrate that if mild modal metasomatism occurs, only one mineral is deposited (e.g., MENZIES and HAWKESWORTH, 1987; NEAL, 1988). This is usually a hydrous mineral, either amphibole or phlogopite. However, MENZIES and WASS (1983) docu-

Table 3: Calculated REE contents of whitlockite equilibrium liquids.

No.	73216 ,36	73216 ,42	14321* ,1211	73216 ,38	14305* ,400	14304 ,86	14305 ,303	14313 ,70	14321 ,1060
Rock	M	M	M	A	A	A	A	A	A
La	928	380	438	396	483	711	586	319	335
Ce	2548	1085	1172	1162	1217	1769	1054	586	588
Nd	1043	558	709	630	648	963	999	608	512
Sm	386	223	188	265	157	300	270	166	148
Eu	—	—	1.6	—	3.7	—	—	—	—
Gd	416	239	155	263	126	271	214	112	119
Dy	269	200	228	184	173	354	261	162	135
Er	149	105	144	74.4	105	232	233	128	114
Yb	200	136	137	74.9	87.6	217	180	90.8	76.6

A = Alkali Anorthosite; B = Basalt; F = Ferroan Anorthosite; G = Alkali Gabbroanorthite; M = Mg Anorthosite.

No.	67975 ,136	67975 ,44Nm	67975 ,44Nf	88105 ,32	88105 ,32	12040	12003	12013,10	14310,20B
Rock	G	G	G	F	F	B	B	B	B
La	717	433	785	284	255	195	382	801	821
Ce	1895	1576	2148	523	462	746	1314	1656	2304
Nd	1141	1247	1327	507	446	708	905	1589	1658
Sm	307	438	390	158	156	156	398	734	830
Eu	8.8	—	—	—	—	—	—	—	—
Gd	217	326	287	135	106	281	359	713	583
Dy	368	558	504	178	152	276	272	298	552
Er	240	450	242	163	129	124	111	219	540
Yb	272	519	237	76.6	55.9	68.1	66.3	242	382

No.	12040 ,39	12036 ,9
Rock	B	B
La	195	242
Ce	710	820
Nd	708	507
Sm	156	127
Eu	—	—
Gd	281	223
Dy	276	189
Er	124	71.1
Yb	88.5	44.2

mented the occurrence of mantle xenoliths from South Australia, which had been modally metasomatized with apatite. These authors postulated that degassing of mantle CO₂ was responsible for causing apatite metasomatism.

Lunar metasomatism will be unlike terrestrial mantle metasomatism because of the lack of OH⁻ and H₂O. Rather,

we envisage a low-viscosity, volatile-rich (e.g., F, Cl) silicate melt as the metasomatic fluid, as evidence of volatiles (especially halogen complexes) is found on the surfaces of glass beads of volcanic origin (e.g., CHOU *et al.*, 1975; MEYER *et al.*, 1975). The basic liquid produced by SLI is a prime candidate because of its low viscosity and halogen-rich nature

Table 4: Selected KREEP/urKREEP REE compositions, liquid/liquid REE Kd's, and the REE contents of basic immiscible melts derived from these KREEPy compositions. The calculated melt compositions are for low-P₂O₅ (first number in each MELT column) and high-P₂O₅ (second number in each MELT column) system liquid/liquid Kd's. Parentheses indicate extrapolated Kd's.

Ref.	1, 2		3	4	5	6	7	
	Kd							
	Low-P ₂ O ₅	High-P ₂ O ₅	KREEP	MELT	KREEP	MELT	KREEP	MELT
La	3.9	13.8	200	780-2760	233	909-3215	110	429-1518
Ce	(4.0)	(14.0)	460	1840-6440	615	2460-8610	280	1120-3920
Nd	(4.2)	(14.4)	270	1134-3888	368	1546-5299	178	748-2563
Sm	4.4	(14.9)	66.0	290-983	98.1	432-1413	48.0	211-715
Eu	(4.5)	(15.1)	3.3	14.9-49.8	1.56	7.02-23.6	3.3	14.9-49.8
Gd	(4.6)	(15.3)	82.8	381-1267	80.1	368-1226	58.0	267-887
Dy	(4.9)	(15.8)	102	497-1612	98.5	483-1556	65.0	319-1027
Er	(5.2)	(16.2)	66.2	343-1072	64.7	336-1048	40.0	208-648
Yb	5.5	16.6	64.0	352-1062	64.3	354-1067	36.0	198-598

MELT = Basic immiscible melt calculate from the KREEP compositions and quoted liquid-liquid Kd's.

References: 1 & 2 = liquid-liquid Kd's of RYERSON and HESS (1980); 3 = urKREEP composition of MORRIS *et al.* (1990); 4 = average calculated urKREEP composition reported by NEAL & TAYLOR (1989); 5 = High-K KREEP of WARREN (1988)

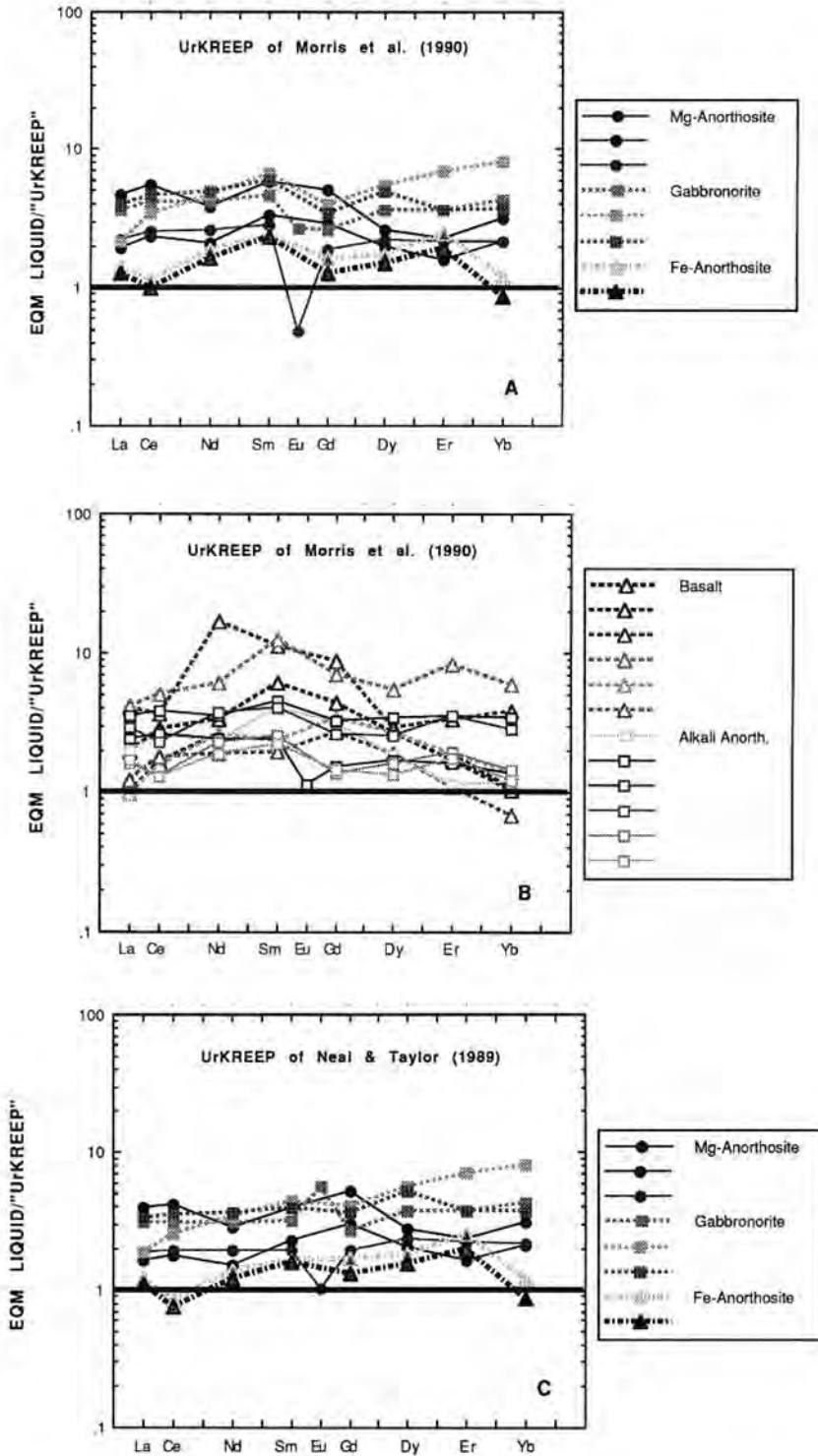


FIG. 6. Whitlockite equilibrium liquids normalized to 3 KREEP or urKREEP compositions. Equilibrium liquids are normalized to: (a, b) the urKREEP composition of MORRIS et al. (1990); (c, d) the urKREEP composition of NEAL and TAYLOR (1989); (e, f) the high-K KREEP of WARREN (1988). Note that the whitlockite equilibrium liquids are more enriched than the respective urKREEP/KREEP compositions (i.e., normalized values are >1). This suggests that the whitlockites did not crystallize directly from urKREEP.

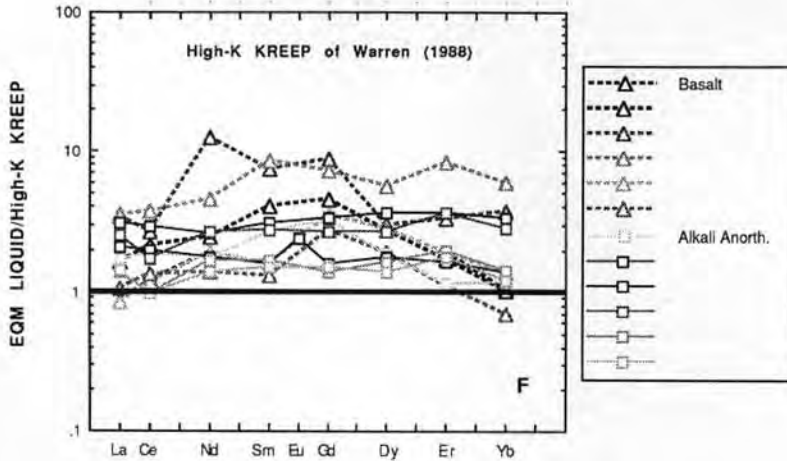
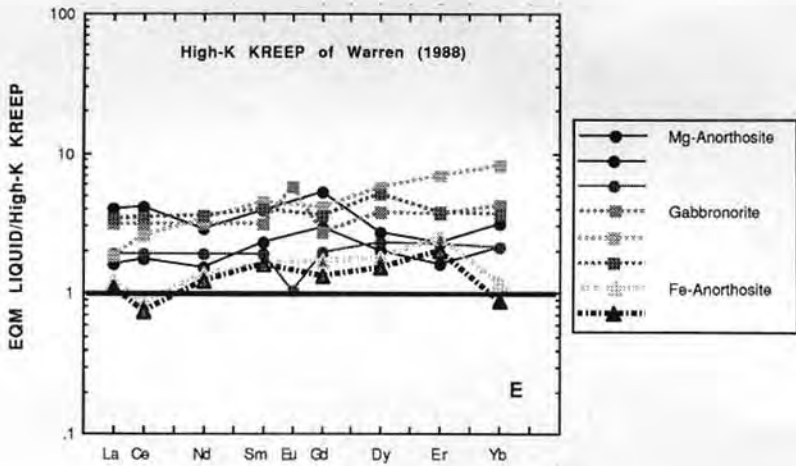
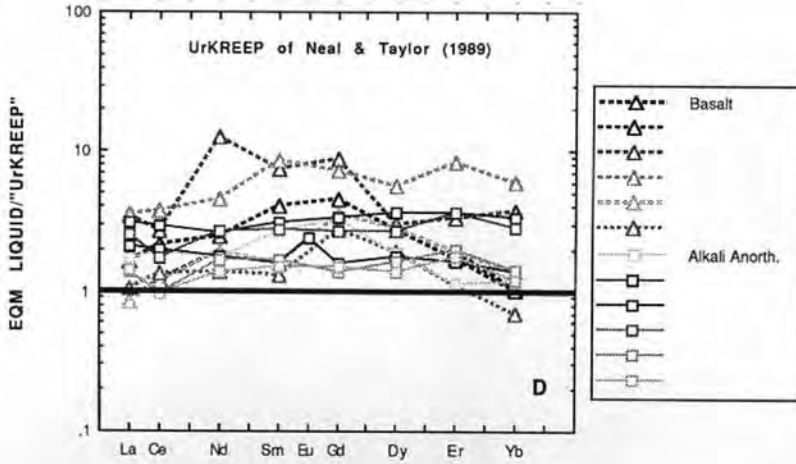


FIG. 6. (Continued)

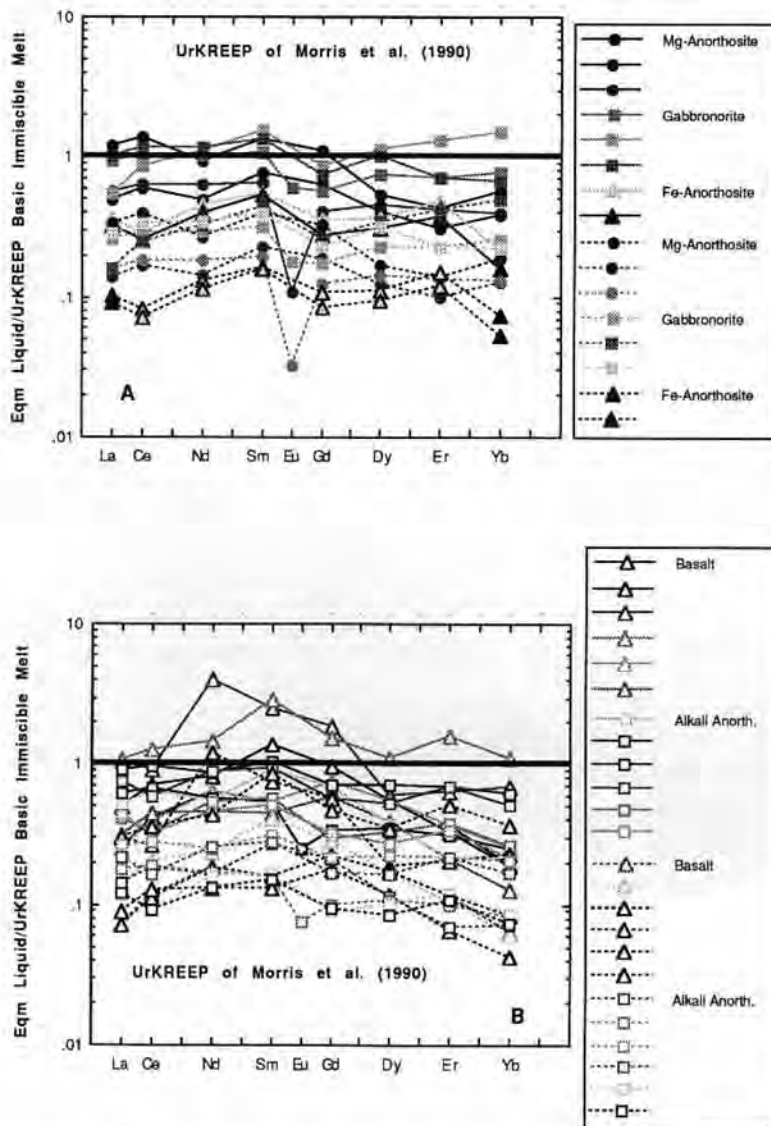


FIG. 7. Whitlockite equilibrium compositions normalized to the basic immiscible melt produced by silicate liquid immiscibility of: (a, b) the urKREEP composition of MORRIS et al. (1990); (c, d) the urKREEP composition of NEAL and TAYLOR (1989); (e, f) the high-K KREEP of WARREN (1988). Note that values are approximately 1 for basic immiscible melts calculated using low- P_2O_5 system liquid/liquid K_d 's (solid lines) and $\ll 1$ for melts calculated using high- P_2O_5 system liquid/liquid K_d 's (dashed lines).

(NEAL and TAYLOR, 1989). After SLI, the granitic melt (K-Fraction) is extremely viscous and will be unable to migrate. Furthermore, this viscous melt will require slow cooling in order for crystals to form. Relative to the K-Fraction, the basic melt (REEP-Fraction) will be able to flow and crystallize comparatively easily. This difference in crystallizing abilities can be seen in the basaltic mesostases, where the REEP-Fraction has crystallized, but the K-Fraction is a quenched glass (e.g., TAYLOR et al., 1971). The viscosity differences between the K- and REEP-Fractions allowed their separation, and post-SLI crystal fractionation of ilmenite and fayalite reduced

the density of the REEP-Fraction, promoting upward migration and metasomatism (NEAL and TAYLOR, 1989). However, crystallization of such phases will push the composition back into the immiscibility gap and more granite will be exsolved (e.g., HESS et al., 1975; RUTHERFORD et al., 1976). Although we do not understand the fluid dynamics of this situation, the two immiscible melts must separate as we find lunar granite (of compositions consistent with formation by SLI) without any ferrobaltic counterpart (RUTHERFORD et al., 1976). It may be that as the basic immiscible melt moves away from the original granitic coun-

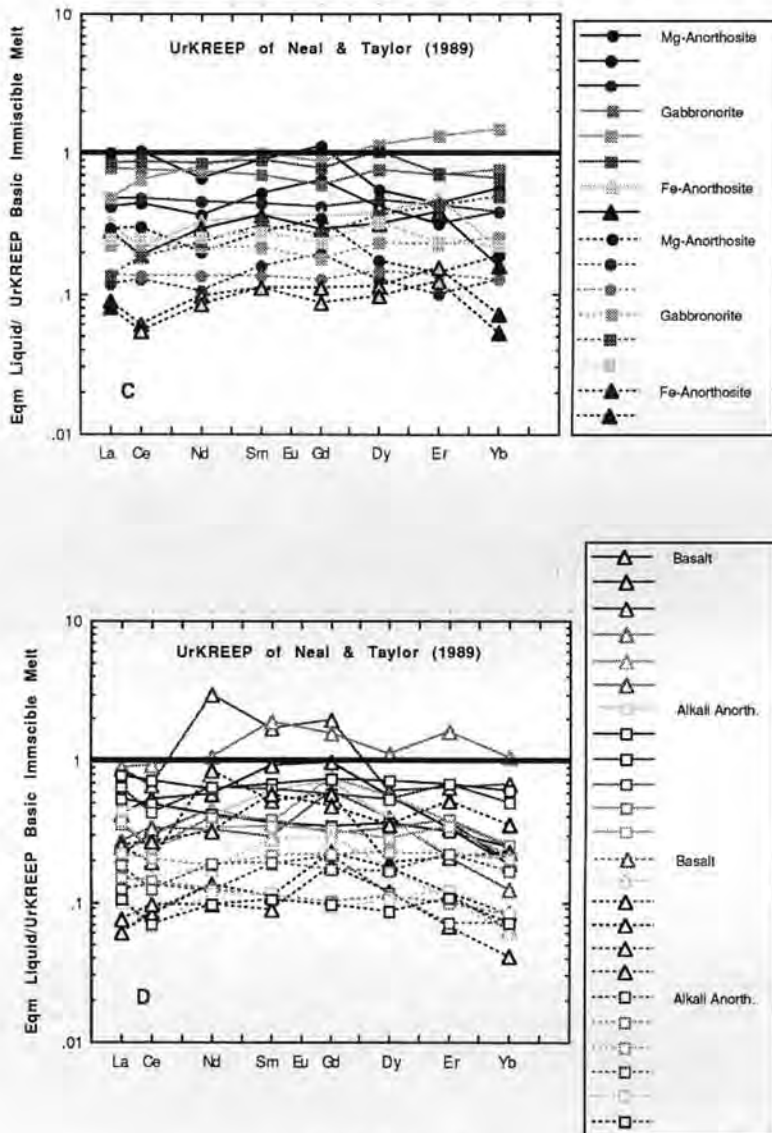


FIG. 7. (Continued)

terpart, it crystallizes ilmenite and fayalite and exsolves more granite while becoming less dense and more enriched in phosphate. This may account for the presence of predominantly whitlockite in the interstices of the certain highland cumulates. RUTHERFORD *et al.* (1976) suggested gravity separation of granitic and ferrobasic immiscible liquids to account for the presence of lunar granite particles without any ferrobasic counterpart in lunar soils and breccias.

HESS *et al.* (1990) have addressed the apatite/whitlockite association in lunar highland cumulates reported by GOODRICH *et al.* (1986) and MARVIN *et al.* (1990). HESS *et al.* (1990) suggested that it is difficult to argue that one or other of these phosphates is the product of open system metasomatism, but agreed that both whitlockite and apatite crys-

tallized from highly evolved melts and suggested that apatite crystallized after whitlockite, possibly in response to changing fluorine fugacities. However, JAMES *et al.* (1987) suggested on the basis of composite phosphate grains within gabbro-norite that either apatite and whitlockite crystallized simultaneously or apatite crystallized before whitlockite.

As noted by LINDSTROM *et al.* (1985, 1991), GOODRICH *et al.* (1986), and HESS *et al.* (1990), whitlockites contain greater REE abundances than apatites, but phosphate-liquid REE partitioning studies demonstrate that coexisting apatite and whitlockite should have identical REE contents (DICKINSON and HESS, 1983; WATSON and GREEN, 1981). Where whitlockite crystallized before apatite in these highland lithologies, it is probably because of low fluorine fugacities in

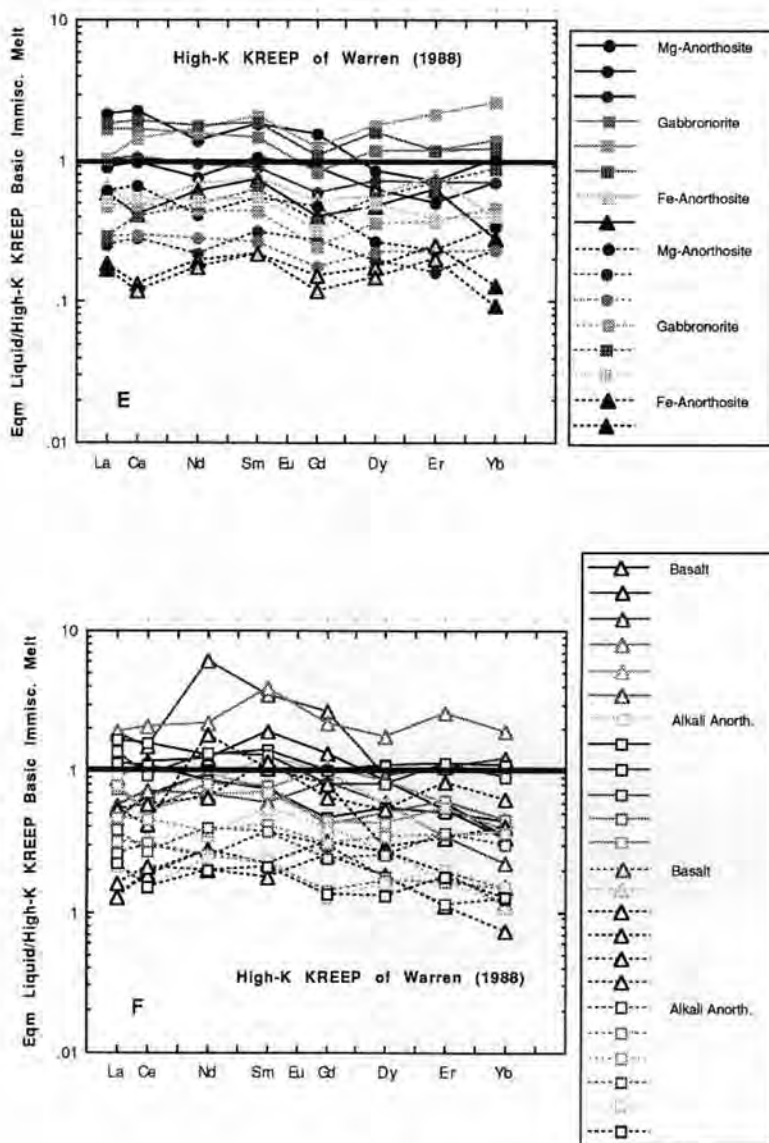


FIG. 7. (Continued)

the REEPy melt (HESS et al., 1990) and the apatite and whitlockite may have similar REE Kd's. If this metasomatic melt could no longer migrate, fluorine could build up due to early whitlockite crystallization which then promotes apatite crystallization. If the melt continued to migrate after whitlockite crystallization, discrete apatites and whitlockites would be produced. As whitlockite crystallized first, the REE abundances in the metasomatic melt will be severely reduced. Therefore, while the later crystallizing apatites would have similar REE-liquid Kd's to apatite, their REE contents will be lower.

The REEPy metasomatic melt has the potential to crystallize ilmenite, plagioclase and possibly K-rich feldspar and even silica (LU et al., 1989). These considerations are con-

sistent with the observations of JAMES et al. (1987) in the gabbronorites, although the timing of apatite and whitlockite crystallization is problematical in this case. Co-crystallization of apatite and whitlockite would suggest that the REE/liquid Kd's would be lower in apatite and higher in whitlockite in order to account for observed REE abundances, which is at variance with some experimental data (DICKINSON and HESS, 1983). Likewise in the case of apatite crystallizing before whitlockite, unless (1) the REE abundances of the parental melt were radically elevated after apatite and prior to whitlockite crystallization, (2) whitlockite and apatite crystallized from separate melts of different REE composition, or (3) the REE Kd's for apatite and whitlockite are not the same.

Experimental evidence to suggest that REE Kd's are greater

in whitlockite than apatite was reported by MURRELL et al. (1984). These authors conducted experiments upon glasses of a diopside, albite, and anorthite composition to which 15% by weight of synthetic chlorapatite or $\text{Ca}_3(\text{PO}_4)_2$ were added. These starting materials were spiked with La, Sm, Yb, and Y oxides to a total REE₂O₃ concentration of approximately 2.5 wt%. Results demonstrated that the crystal/liquid REE partition coefficient for whitlockite was 4–5 times greater than for apatite but the Kd patterns for both phosphates were parallel. The LREEs have a higher D (La ~ 5.0 for whitlockite; La ~ 1.3 for apatite) than the HREEs (Yb ~ 1.8 for whitlockite; Yb ~ 0.6 for apatite). This dichotomy between REE Kd's for apatite and whitlockite may explain the apparent co-crystallization of apatite and whitlockite observed by JAMES et al. (1987), yet whitlockite contains the higher REE abundances. If the absolute REE Kd values reported by MURRELL et al. (1984) are correct, then only a REEPy melt derived from a high-P₂O₅ system would contain high enough REE abundances to produce observed levels in whitlockites. Clearly, careful examination of REE partitioning between these phosphates and melt are required.

CONCLUSIONS

On the basis of calculate whitlockite/liquid REE Kd's, the only liquid containing sufficient REEs (and Ca and P) to account for lunar whitlockite compositions is a basic immiscible melt (REEP-Fraction) produced from urKREEP/KREEP undergoing SLI. This is evident by comparison of the calculated REE contents of whitlockite equilibrium liquids and basic immiscible melts from KREEP/urKREEP. Even in basaltic mesostases, the process of SLI appears to be important in the paragenesis of whitlockite. The nature and composition of whitlockite in the lunar highlands, and lack of associated minerals, is not consistent with the *in situ* crystallization of the intercumulus liquid, even if it did undergo SLI; the interstitial mineral assemblage in these rocks does not contain a sufficient number of phases. However, the interstitial mineral assemblage which is present is consistent with metasomatism by comparison with terrestrial examples, although the mineral compositions are different. No hydrous minerals are present on the Moon, so the metasomatic fluid is a low viscosity silicate melt, again consistent with the basic immiscible melt from urKREEP/KREEP. Post-SLI fractionation of ilmenite/fayalite from the REEP-Fraction reduces density and, coupled with low viscosity, allows percolation of and metasomatism by this melt. Changing fluorine fugacities as this metasomatic melt evolves may facilitate the eventual crystallization of apatite (after whitlockite). This apatite may form epitaxial overgrowths on the early whitlockite (if the metasomatic melt cannot migrate further) or form discrete apatites and whitlockites in the same rock, if percolation can continue. The early crystallization of whitlockite increases the halogen content of the residual melt, but reduces the REE contents. Later crystallization of apatite, while containing similar REE Kd's to whitlockite, will contain lower REE abundances reflecting the composition of the residual melt. However, the experimental results of MURRELL et al. (1984) combined with the observations of JAMES et al.

(1987) indicate that at least in some cases apatite co-crystallizes with whitlockite and has lower REE Kd's. Apatite which appears to form before or with whitlockite may be accommodated within this metasomatic model once accurate phosphate/liquid partition coefficients have been determined. Even so, a melt highly enriched in the REEs (and P) is required to facilitate whitlockite crystallization. Our model suggests that such a melt composition is consistent with that proposed for the REEP-Fraction and is consistent with a metasomatic origin.

The recognition of lunar metasomatic indicators is difficult because of the lack of hydrous minerals. However, the process of silicate liquid immiscibility produces a silicate melt which is ideal for lunar metasomatism. This metasomatic melt, derived from KREEP/urKREEP or equivalent compositions, can account for the presence of whitlockites of similar compositions in a variety of lunar basalts and cumulate rocks, and the apparent apatite/whitlockite associations.

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