American Mineralogist, Volume 93, pages 488–491, 2008

LETTER

Rhönite in Luna 24 pyroxenes: First find from the Moon, and implications for volatiles in planetary magmas

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ABSTRACT

Grains of rhönite have been discovered in magmatic inclusions in augite grains of the lunar regolith from Mare Crisium, returned to Earth by the Russian Luna 24 spacecraft. These rhönite grains are up to 8 μm long, pleochroic from tan to dark brown, and associated with ulvöspinel and silica-rich glass. Electron microprobe analysis gives a composition near end-member ferroan rhönite: $(Ca_{1.9}Mn_{0.0}Na_{0.1})(Fe^{2+}_{4.5}Mg_{0.1}Al_{0.3}Cr_{0.0})Ti_{1.0}(Si_{4.0}Al_{2.0})O_{20}$. The Raman spectrum of these grains is like those of terrestrial rhönites, and distinct from titanian amphiboles. Compositionally, rhönite plus silica plus water or halogens (F, Cl) is equivalent to titanian amphibole plus pyroxene, so the presence of rhönite in lunar basaltic rock is consistent with the known low abundance of volatiles in the Moon. When calibrated, mineral reactions involving rhönite and titanian amphibole may provide quantitative constraints on fugacities of water, F, and Cl in basaltic magmas from the Moon and other planetary bodies.

Keywords: Lunar and planetary studies, mare basalt, Luna 24, igneous petrology, moon, basalt, melt inclusion, petrography, rhönite

INTRODUCTION

Compared to the Earth, the Moon is strongly depleted in volatiles, and its interior is generally considered anhydrous (Heiken et al. 1991; Spudis 2001). The internal volatile content of the Moon remains a critical control on its petrogenesis and is being reassessed in light of current interest in the Moon (Saal et al. 2007). One tool in this assessment is the investigation of minerals that could contain volatile elements.

Amphibole is one such volatile-bearing mineral, as it can contain H_2O (as OH), F, and Cl. Amphiboles were reported in several lunar rocks (Gay et al. 1971; Mason et al. 1972), but these finds have not been verified. A report of amphibole in Luna 24 regolith olivine grains (Laz'ko et al. 1980) was the impetus for this study. I have not found amphibole, but have found the first lunar occurrence of the anhydrous mineral rhönite (Treiman 2007), which has a composition similar to that of kaersutite amphibole. The rhönite is found in magmatic inclusions in pyroxene—the same setting for kaersutite amphibole in the shergottite martian basalts (Treiman 1985, 1997, 1998).

SAMPLES AND METHODS

Grain mount thin sections of Luna 24 regolith were examined optically at the ARES thin section library, Building 31, Johnson Space Center. Selected thin sections were borrowed for further analysis via a request to the CAPTEM lunar allocation subcommittee. The largest rhönite grain is in thin section 24105,15—fortunately, it is exposed at the surface of the thin section. The allocated sections were examined optically at the Lunar and Planetary Institute, and then subjected to Raman and electron microprobe analyses at JSC.

Spot micro-Raman analyses were taken with the Horiba HR-LabRam Raman microscope at ARES (JSC) using red He-Ne laser light. The laser light was focused through a long-focal length objective lens to a spot of ~3 μm diameter on the thin section surface. We obtained several Raman spectra of the rhönite grain in 24105,15, and of the adjacent surrounding pyroxene, siliceous glass, and ulvöspinel. Raman scans were from 300 to 1100 cm^{-1}.

Chemical analyses and the back-scattered electron image were obtained by electron microprobe (EMP), using the Cameca SX-100 at ARES (JSC), 15 kV accelerating potential, beam current 10 na, beam diameter 1 μm. For quantitative element analyses, most elements were standardized against synthetic crystalline oxides. Other standards were orthoclase for K, oligoclase for Na, fluorite for F, and tugtupite for Cl. Data were reduced in the Cameca PAP routine. Analyses for siliceous glass are not considered quantitative, because standards did not include glasses, and because the focused beam likely caused significant volatile loss and element mobility.

RESULTS

The L24 regolith includes mineral fragments, rock fragments, and glasses (Coish and Taylor 1978). Most mineral fragments are pyroxenes and olivine, which are interpreted as fragments of crystalline basalts and gabbros emplaced in the Mare Crisium basin. Many of these pyroxene grains contain glassy inclusions (with or without crystalline phases) that are inferred to be melt inclusions—remnant of magma trapped in crystals as they grew (Roedder and Weiblen 1978).

Melt inclusions in many pyroxene grains contain tabular brown crystals—approximately 20 such grains have been found in the three investigated grain-mount sections (L24105,15; L24149,41; L24174,64). These grains may have been noted before by Roedder and Weiblen (1978, p. 497) as "… roughly tabular, transparent, yellow-brown daughter crystal[s], presumably a spinel…." The two largest brown crystals are ~10 μm in length, and one in L24105,15 is cut by the thin section surface (Fig. 1); all optical and chemical data are for that grain. The

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0003-004X/08/0203–488$05.00/DOI: 10.2138/am.2008.2781

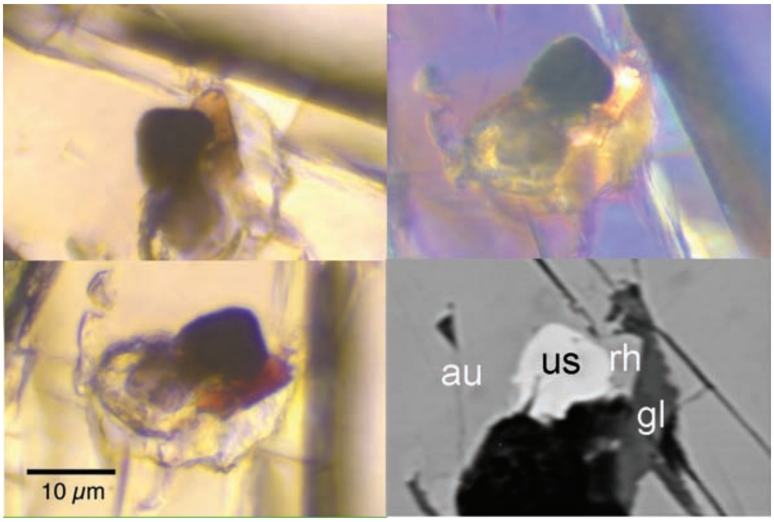

Figure 1. The largest L24 rhönite grain, in a magmatic inclusion within a grain of augite; all at same scale. Images on left (top and bottom) are in plane light, rotated 90° to each other, to show pleochroism. Rhönite is brown, ulvöspinel is opaque, and the colorless material between rhönite and augite is silica-rich glass. Image on top right is in crossed polarized light, with host augite near extinction, showing the birefringence of the rhönite. Image at bottom right is by back-scattered electrons (BSE), showing rhönite (rh), ulvöspinel (us), glass (gl), and augite (au).

Table 1. EMP chemical compositions of rhönite and adjacent materials in L24105,15

	rhönite average	1σ	pyroxene	ulvöspinel	glass
SiO_2	27.40	±0.27	45.81	0.07	*66.9*
TiO_2	8.99	±0.28	1.17	30.90	*0.1*
Al_2O_3	13.27	±0.16	3.22	5.12	*14.0*
Cr_2O_3	0.07	±0.01	0.05	0.56	*0.0*
FeO	37.21	±0.11	37.04	60.35	*2.8*
MnO	0.16	±0.01	0.46	0.35	*0.1*
MgO	0.56	±0.03	2.00	0.09	*0.1*
CaO	11.96	±0.50	11.01	0.32	*8.2*
Na_2O	0.36	±0.36	0.06	0.00	*0.9*
K_2O	0.01	±0.01	0.00	0.00	*1.9*
Cl	<0.05		–	–	*0.0*
F	<0.01		–	–	*<0.1*
sum	99.99	±0.26	100.83	97.94	*95.1*
Fe/Mg	37		10	374	–
Fe/Mn	237		79	170	–

Notes: Rhönite values are the average of three individual analyses, all within anaytical uncertainty of each other. Low total for ulvöspinel reflects a small proportion of ferric iron. Glass analysis is semi-quantitative, as indicated by italics—low total reflects use of non-ideal standards, and losses and mobility of alkalis; it *cannot* be construed to indicate an abundance of volatiles.

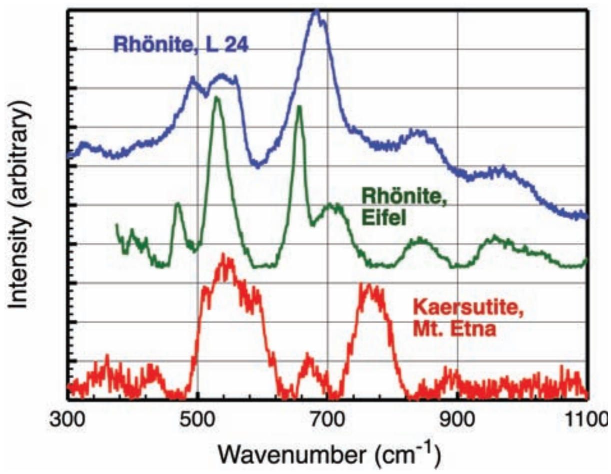

Figure 2. Raman scattering spectrum of the L24 rhönite grain (Fig. 1), compared with spectra of a terrestrial rhönite and a terrestrial kaersutite amphibole (RRUF spectra 060316 and 070128, respectively, unoriented grains, 785 nm excitation; Downs 2006). Intensity axis scaled arbitrarily for presentation.

brown grains are optically anisotropic, length-slow, pleochroic from pale greenish brown to reddish brown (in the slow direction), and with inclined extinction (Z∧c = 58° for the grain of Fig. 1). These optical properties are not consistent with previously reported lunar minerals, but are consistent with kaersutite amphibole (as is found in similar magmatic inclusions in martian meteorite pyroxenes; Treiman 1985) and with rhönite (Anthony et al. 2003). Other minerals in the melt inclusions are ulvöspinel, silica-rich glass (Table 1), and Fe-sulfide (not analyzed). Iron metal was not noted.

Fortuitously, the largest grain is exposed at the surface of the thin section, and is big enough (~8 μm long) for multiple chemical analyses by EMP. The EMP analyses are all within analytical uncertainty of each other, so an average is given in Table 1. The brown grain contains ~25% SiO_2—far too little silica to be kaersutite or other amphibole, but consistent with the mineral rhönite (Söllner 1907). The EMP chemical analysis normalizes properly for rhönite stoichiometry as $(Ca_{1.88}Mn_{0.02}Na_{0.09})(Fe^{2+}_{4.56}Mg_{0.12}Al_{0.31}Cr_{0.01})Ti_{0.99}(Si_{4.02}Al_{1.98})O_{20}$, which is close to the ideal formula: $Ca_2M^{2+}_5Ti(Al_2Si_4)O_{20}$. Ferric iron is not required for charge balance in the L24 rhönite; in fact, its chemical normalization has a slight excess of cation charge, which could suggest the presence of some Ti^{3+} (as reported in some asteroidal rhönites, Nazarov et al. 2000).

The grain's identity as rhönite is confirmed by its Raman scattering spectrum, Figure 2. Its Raman spectrum shares major scattering peaks with a literature rhönite spectrum, of a grain from the Eifel district, Germany (Downs 2006). Specifically, both rhönite spectra have Raman scattering peaks near 540, 670, 720, 840, and 980 cm^{-1} (Fig. 2). Differences between the rhönite spectra can be ascribed to their widely differing chemical compositions (ferric absent for L24; ferric rich for Eifel; Rondorf 1989). Raman spectra of kaersutite amphiboles are completely distinct from those of rhönite (Fig. 2).

DISCUSSION

Rhönite

Rhönite is a mineral in the aenigmatite group of branched single-chain silicates (Cosca et al. 1988; Kunzmann 1999), which accommodate a wide range of elemental substitution. Rhönite is a widespread but uncommon accessory mineral in terrestrial basaltic rocks, particularly silica-undersaturated varieties, and is commonly rich in ferric iron. In basalts, rhönite occurs as microphenocrysts, and as larger grains in late-stage differentiates (e.g., Johnston and Stout 1985; Cosca et al. 1988; Grapes et al. 2003; Nédli and Tóth 2003; Corsaro et al. 2007). Rhönite is also found occasionally in magmatic inclusions, as reported here for the Luna 24 sample (Kothay et al. 2003; Jannot et al. 2005; Timina et al. 2006). Rhönite also is present among the breakdown products (in "opacite" rims) of kaersutite and related amphiboles (O'Connor et al. 1996; Monkawa et al. 2003; Grapes et al. 2003; Kovács et al. 2004; Alletti et al. 2005; Corsaro et al. 2007).

Rhönite and related minerals are present in a few extraterrestrial samples, notably from calcium-aluminum-rich inclusions (CAIs) in carbonaceous chondrites (e.g., Fuchs 1971; Simon et al. 1999; Nazarov et al. 2000). Aenigmatite has been found in chondritic breccias (Ivanov et al. 2003; Bischoff et al. 2006), and its original source may also have been CAIs or related material. Rhönite might also be expected in angrite meteorites, asteroidal basalts rich in calcium and aluminum; it has recently been found in one (NWA 4950; Kuehner and Irving 2007), and has been produced in laboratory experiments on angrite compositions (Lofgren et al. 2006). A singular occurrence of rhönite in a ureilite meteorite is as small euhedra in an Al-rich glassy enclave, which could have affinities to the angrite basalts (Warren et al. 2006).

Geologic setting

The L24 rhönite grains are all in multi-phase inclusions in augite pyroxenes. Many of the augite grains are distinctly zoned, with an inclusion-free pale core and an inclusion-rich pink-colored rim. All pink-colored pyroxenes contain multiphase inclusions, but not all such pyroxenes contain rhönite-bearing inclusions. The pyroxene adjacent to the rhönite grain of Figure 1 is ferrohedenbergite ($Wo_{26}En_{06}Fs_{68}$) with a lunar Fe/Mn ratio (Table 1; Papike et al. 2003), and is rich in Al and Ti and poor in Cr. Formula normalizations show slight charge excesses, which could indicate that some Ti in the augite is Ti^{3+} rather than Ti^{4+}. Pyroxenes like those hosting the rhönite-bearing inclusions are common in the Luna 24 regolith in fragments of very-low-titanium (VLT) basalt and as isolated grains ascribed to VLT basalt or VLT-derived ferro-gabbro (Bence et al. 1977; Coish and Taylor 1978; Papike and Vaniman 1978; Roedder and Weiblen 1978; Lu et al. 1989).

SIGNIFICANCE

This is the first report of rhönite from a lunar sample. Beyond being a mineralogical oddity, this find shows that the returned lunar samples have not been "mined out" scientifically—much remains to be discovered and learned with modern techniques and new minds!

The petrologic significance of this rhönite occurrence is its implications for volatiles in lunar magmas. Lunar magmas have been considered essentially anhydrous (e.g., Heiken et al. 1991; Spudis 2001), but some lunar basalt glasses contain tens of ppm H_2O (Saal et al. 2007) and some reports of amphiboles suggested abundant water or other volatiles (Gay et al. 1971; Mason et al. 1972). The find here of rhönite is consistent with a relatively low volatile content in the Luna 24 basalts, because titanian amphibole would be expected in "volatile-rich" systems. This inference is exemplified in the reaction

$$\underset{\text{titano-tschermakite}}{Ca_2(M^{2+}_4Ti)(Al_2Si_6)O_{22}(OH)_2} + \underset{\text{pyroxene}}{M^{2+}SiO_3} = \underset{\text{rhönite}}{Ca_2M^{2+}_5Ti(Al_2Si_4)O_{20}} + 3SiO_2 + H_2O$$

where M^{2+} represents Fe^{2+} and Mg^{2+}, titano-tschermakite is a model for kaersutitic amphibole, and the silica and water might reside in siliceous melt (Table 1). Comparable reactions can be written for fluor- and chlor-amphiboles. The implication of this reaction is that rhönite should form only in "volatile-poor" systems. This reaction is realized in some cases, where rhönite occurs as a dehydration/devolatilization product of titanian amphibole (O'Connor et al. 1996; Monkawa et al. 2003; Grapes et al. 2003; Alletti et al. 2005; Corsaro et al. 2007). Unfortunately, the locations of these and other reactions involving rhönite are poorly known (e.g., Boivin 1980; Cosca et al. 1988; Kunzmann 1999), so the phrase "volatile-poor" cannot yet be quantified.

On the other side of this equation, kaersutite amphibole (Ti-rich) is found in magmatic inclusions in many martian basaltic meteorites (Floran et al. 1978; Treiman 1985, 1997, 1998; Ikeda 1997, 1998; Mikouchi and Miyamoto 2000; Sautter et al. 2006). These kaersutites have been used to suggest that the martian basalts contained abundant water (Johnson et al. 1991; McSween and Harvey 1993); however, these amphiboles are oxy-kaersutites and contain nearly no OH (Watson et al. 1994). The oxy-kaersutite in the martian meteorites could have formed by dehydrogenation/oxidation of hydrous kaersutite or could be a primary igneous composition (Popp et al. 1995; Mysen et al. 1998; McCubbin et al. 2006). If the latter, it is not clear why oxy-kaersutite might form instead of a Fe^{3+}-bearing rhönite. So, knowing the relative stabilities of titanian amphiboles and rhönite-group minerals would provide important constraints on the volatile contents of lunar, martian, and some terrestrial magmas (Cosca et al. 1988).

ACKNOWLEDGMENTS

I am grateful to L. Le (Jacobs Sverdrup, JSC) for technical assistance with EMP and Raman analyses, to A.J. Irving for sharing pre-publication data, and to E.J. Essene for intellectual mentorship and stimulating discussions. The RRUF mineral spectroscopy database (Arizona State University) was the source of reference mineral spectra. GoogleScholar was invaluable in searching for literature on rhönite. Reviews by J. Papike and M. Rutherford are deeply appreciated. Supported by NASA MFR grant NNG06GH29G. Lunar and Planetary Institute contribution no. 1362.

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MANUSCRIPT RECEIVED SEPTEMBER 6, 2007
MANUSCRIPT ACCEPTED OCTOBER 19, 2007
MANUSCRIPT HANDLED BY BRYAN CHAKOUMAKOS