

# Manganese-rich olivines: Identification from spectral reflectance properties

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**Abstract.** Reflectance spectra of manganese-rich olivines were examined to determine which spectral features allow these minerals to be distinguished from forsteritic-fayalitic olivines. The results indicate that manganese-rich olivines can be distinguished on the following bases:  $\text{Fe}^{2+}$  M2 absorption band depths are reduced relative to  $\text{Fe}^{2+}$  M1 depths, the wavelength position of the  $\text{Fe}^{2+}$  M2 absorption band is shifted to longer wavelengths ( $> 1.08 \mu\text{m}$ ) for olivines containing between  $\sim 10$  and 70 mol %  $\text{Mn}_2\text{SiO}_4$ , and the presence of an isolated  $\text{Mn}^{2+}$  transition band near  $0.4 \mu\text{m}$ . The absence of  $\text{Fe}^{2+}$  spin forbidden features in the visible wavelength region is not a reliable method for identifying manganese-rich olivines.

## 1. Introduction and Background

King and Ridley [1987] conducted a comprehensive examination of the spectral reflectance properties of a suite of olivine samples spanning a range of compositions. Many of their samples contained, at most, a few weight percent  $\text{MnO}$ , and they interpreted the spectral effects of manganese on the basis of these spectra as well as earlier transmission spectra of compositionally diverse olivines [Burns, 1970; Burns and Huggins, 1972; Burns *et al.*, 1972].

Reflectance spectra of some Mn-rich olivines were acquired in order to determine whether the interpretations of the effects of Mn on the reflectance spectra of olivines as suggested by King and Ridley [1987] were correct and whether manganese-rich olivines exhibit unique and diagnostic spectral properties.

A number of studies have indicated that  $\text{Mn}^{2+}$  exhibits a strong preference for the M2 structural site in olivine [Francis and Ribbe, 1980; Miller and Ribbe, 1985]. This is the same site, which when occupied by  $\text{Fe}^{2+}$ , gives rise to an absorption feature centered near  $1.05 \mu\text{m}$ . This site preference has important implications for distinguishing Mn-rich from Mn-poor olivines.

Cation substitutions of relevance to this study involve Mg with  $\text{Fe}^{2+}$  and  $\text{Fe}^{2+}$  with Mn. Mn does not appreciably substitute for Mg. The pure end-members are forsterite (Mg), fayalite (Fe), and tephroite (Mn). Olivines with compositions intermediate between fayalite and tephroite are termed knebelite [Deer *et al.*, 1966]. Small replacements of Fe and Mn by Zn also occur in some tephroites. This nomenclature is used throughout this paper.

Manganese-rich olivines are most commonly associated with iron-manganese ore deposits and skarns. Manganiferous ore bodies are found in a wide range of environments, many of Archean age [e.g., Evans, 1980]. The nature of some types of manganese deposits has important implications for extraterrestrial exploration. Detection of manganiferous deposits on Mars would be strong evidence for the operation of processes and environments not currently observed, such as deep weathering (lateritization), shallow marine emplacement, or contact metamorphism of carbonates.

If manganiferous olivines do exhibit unique spectral properties, it may be possible to detect such deposits spectroscopically. In the case of carbonate detection, it should be noted that olivines are easier to detect spectroscopically than carbonates because the diagnostic absorption bands of olivines occur at shorter wavelengths than carbonates ( $1 \mu\text{m}$  versus  $> 1.6 \mu\text{m}$  [Gaffey, 1986; King and Ridley, 1987]). However, extraterrestrial occurrences of manganese-rich olivines have not been detected to date, either spectroscopically or directly [e.g., Basaltic Volcanism Study Project, 1981].

## 2. Experimental Procedure

A suite of olivines were acquired as part of an ongoing study of the spectral properties of mafic silicates. All of the olivine samples were characterized by electron microprobe and X ray diffraction analysis (Table 1) and prepared for spectral measurements as outlined in Cloutis and Gaffey [1991a]. OLV005 (Forsterite1) is a forsterite from St. John's Island, Egypt, OLV007 (Forsterite2) is a forsterite from Palumbo Farm, Villafranca, Cecchina, Italy, OLV023 (Tephroite) is a tephroite from Sterling Hills, New Jersey, and OLV105 (Knebelite) is a knebelite from the Highland Bell Mine, Beaverdell, British Columbia, Canada. Reflectance spectra of both  $< 45 \mu\text{m}$  and  $45\text{--}90 \mu\text{m}$  size separates were measured at the RELAB spectrometer facility at Brown University [Pieters, 1983; RELAB, 1996] relative to halon at  $i=30^\circ$ ,  $e=0^\circ$  and 5 nm resolution.

## 3. Results

### 3.1. Effect of Mn on $\text{Fe}^{2+}$ Absorption Band Position

Iron-bearing olivines exhibit a major absorption feature near  $1 \mu\text{m}$  attributable to ferrous iron. This feature is resolvable into three partially overlapping absorption bands [Burns, 1970; Sunshine and Pieters, 1990]. The center band is the most intense and is due to  $\text{Fe}^{2+}$  located in the M2 crystallographic site, while the two outer bands are due to  $\text{Fe}^{2+}$  in the M1 crystallographic site. Manganese shows a marked preference for the slightly larger M2 site because  $\text{Mn}^{2+}$  has a larger octahedral ionic radius than  $\text{Fe}^{2+}$ . With increasing Mn content, the wavelength position of the  $\text{Fe}^{2+}$  M2 band moves to longer wavelengths in transmission spectra [Burns, 1970, 1993].

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Table 1. Composition of Selected Olivines

	OLV005 Forsterite1	OLV007 Forsterite2	OLV023 Tephroite	OLV105 Knebelite
SiO <sub>2</sub>	40.97	41.72	28.83	30.19
FeO	9.59	3.12	2.24	46.86
Fe <sub>2</sub> O <sub>3</sub>	n.d.	n.d.	n.d.	n.d.
MgO	49.64	54.65	3.41	1.36
MnO	0.09	0.19	56.90	20.69
ZnO	<0.01	0.00	5.94	0.07
NiO	0.32	0.01	0.25	0.13
CaO	0.00	0.63	0.56	0.04
TiO <sub>2</sub>	<0.01	0.00	0.08	0.02
Cr <sub>2</sub> O <sub>3</sub>	0.00	<0.01	0.04	0.01
CoO	0.06	0.01	0.12	0.11
V <sub>2</sub> O <sub>5</sub>	0.00	0.00	0.07	0.01
K <sub>2</sub> O	0.00	0.00	0.02	0.00
Na <sub>2</sub> O	0.00	0.00	0.59	0.00
Al <sub>2</sub> O <sub>3</sub>	0.00	0.00	<0.01	0.00
Total	100.67	100.33	99.05	99.49
Mg	89.8	95.9	8.3	3.4
Fe <sup>2+</sup>	9.7	3.1	3.0	66.4
Mn	0.1	0.2	78.2	29.7

OLV023 was acquired from the Smithsonian Museum of Natural History [MNH C2824]; n.d., not determined.

The reflectance spectra of the two Mn-rich olivines, tephroite and knebelite, (Figure 1) indicate that the Fe<sup>2+</sup> absorption bands in tephroite (OLV023) may become undetectable, similar to the results of *Burns* [1970]. The knebelite (OLV105), which contains more iron and less manganese than the tephroite, displays a resolvable Fe<sup>2+</sup> absorption feature. The position of the band minimum for the 45-90 μm sized sample is 1.10 μm. The band minimum is expected to be near 1.085 μm on the basis of fayalite content or 1.060 μm on the basis of forsterite content, using the calibration of *King and Ridley* [1987]. Thus the presence of

significant amounts of Mn moves the Fe<sup>2+</sup> absorption band minimum to longer wavelengths in reflectance spectra, as expected [*Burns*, 1970].

The transmission spectra of *Burns* [1970] can be used to calculate the Mn content required to shift the wavelength position of the Fe<sup>2+</sup> M2 band outside the range occupied by forsteritic-fayalitic olivines. These data have been replotted on the basis of both fayalite and tephroite content (Figure 2). Extrapolation of the tephroite trend suggests that olivines with an Mn<sub>2</sub>SiO<sub>4</sub> content greater than ~10 mol % will exhibit spectra with an Fe<sup>2+</sup> M2 absorption band whose wavelength position falls outside the range occupied by forsterite-fayalite. The available data also suggest that when Mn<sub>2</sub>SiO<sub>4</sub> content exceeds some value between 70 and 78 %, the intensity of the Fe<sup>2+</sup> absorption bands will be reduced to the point that they do not appear as readily resolvable features. Thus olivines containing Mn<sub>2</sub>SiO<sub>4</sub> in the range ~10-70 mol % should be spectrally distinct and Mn<sub>2</sub>SiO<sub>4</sub> content may be quantifiable when more data become available to better define the spectral-compositional relationships.

### 3.2. Effect of Mn on Fe<sup>2+</sup> Absorption Band Shape

To better illustrate the effects of Mn on the reflectance spectra of olivines, the major Fe<sup>2+</sup> absorption band region reflectance spectra of two olivines with similar iron contents (Forsterite2: OLV007, and tephroite: OLV023) are shown in Figure 3. The major compositional difference is in manganese (and magnesium) contents (Table 1).

The two spectra differ quite dramatically in appearance. While the Forsterite2 spectrum exhibits a "classical" forsterite-fayalite absorption feature with a prominent Fe<sup>2+</sup> M2 absorption band flanked by the less intense Fe<sup>2+</sup> M1 bands, the tephroite spectrum does not. Since Mn preferentially occupies the M2 crystallographic site, the similar iron contents suggest that, at a minimum, the tephroite spectrum should exhibit two Fe<sup>2+</sup> M1 bands which are at least as deep as the same bands in the Forsterite2 spectrum. The reason for their absence is not fully understood although a transmission spectrum of a tephroite with a similar iron content (1.67% FeO) is also featureless in this wavelength region [*Burns*, 1970].

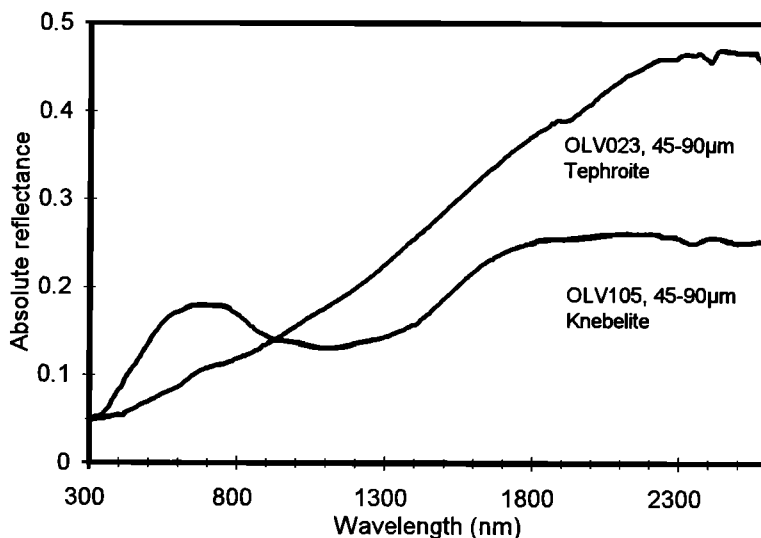


Figure 1. Reflectance spectra of two manganese-rich olivines: tephroite (OLV023; Mg<sub>8.3</sub>Fe<sub>3.0</sub>Mn<sub>78.2</sub>) and knebelite (OLV105; Mg<sub>3.4</sub>Fe<sub>66.4</sub>Mn<sub>29.7</sub>). See Table 1 for complete compositional data.

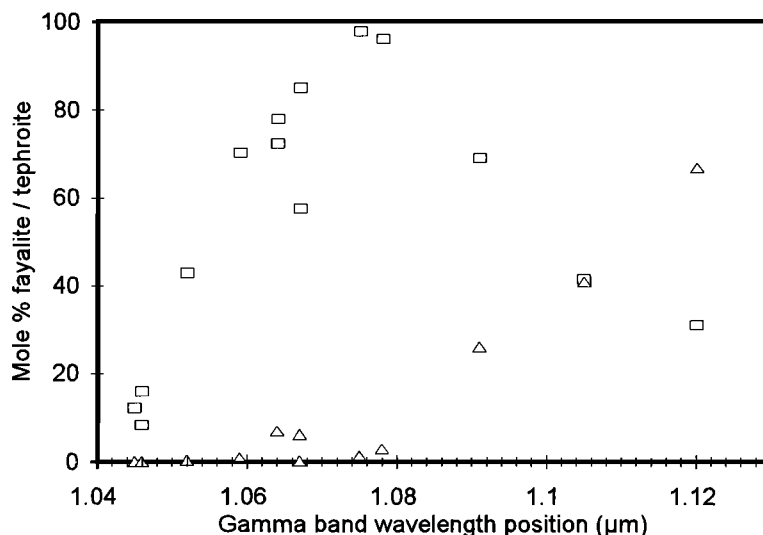


Figure 2. Data on wavelength position of  $\text{Fe}^{2+}$  M2 site  $\gamma$  band from Burns [1970] olivine transmission spectra plotted as a function of iron (fayalite;  $\text{Fe}_2\text{SiO}_4$ ) content (squares) and manganese (tephroite;  $\text{Mn}_2\text{SiO}_4$ ) content (triangles). With one exception (at 1.064  $\mu\text{m}$ ) each square and triangle is vertically paired.

The knebelite spectrum can also be analysed in terms of existing information. It exhibits absorption features consistent with transmission spectra of compositionally similar tephroites, particularly a more subdued  $\text{Fe}^{2+}$  M2 central band than manganese-poor olivines (Figure 4). Its band depth relative to a straight line continuum (20%) is similar to that of Forsterite2 (19%) in spite of the large difference in iron content. Extrapolating from available olivine reflectance spectra, the band depth of the knebelite is at least a factor of 2 less intense than would be expected on the basis of its iron content and grain size.

### 3.3. Effect of Mn on $\text{Fe}^{2+}$ Spin Forbidden Bands

A number of minor absorption bands have been detected in the visible wavelength region of olivine transmission spectra and attributed to  $\text{Fe}^{2+}$  spin-forbidden transitions [e.g., Burns, 1970].

The wavelength positions of the bands attributed to this mechanism by various investigators are provided in Table 2. These values have been provided uncritically, and it should be noted that the assignments of some of these bands are somewhat contentious [e.g., Vaughan and Burns, 1977]. King and Ridley [1987] suggested that the absence of these bands could be used to distinguish manganese-rich from iron-rich olivines.

Comparisons of the reflectance spectra of different size fractions of various olivines suggests that this may be an oversimplification. The reflectance spectra presented by King and Ridley [1987] show that these absorption features are most prominent in the larger grain size fractions and may be nearly undetectable in powdered samples which have been sieved to  $<60 \mu\text{m}$ . This effect is apparent in the visible region reflectance spectra of Forsterite1 (Figure 5), a typical low manganese, forsteritic olivine (Table 1). Absolute reflectance values are similar for both the

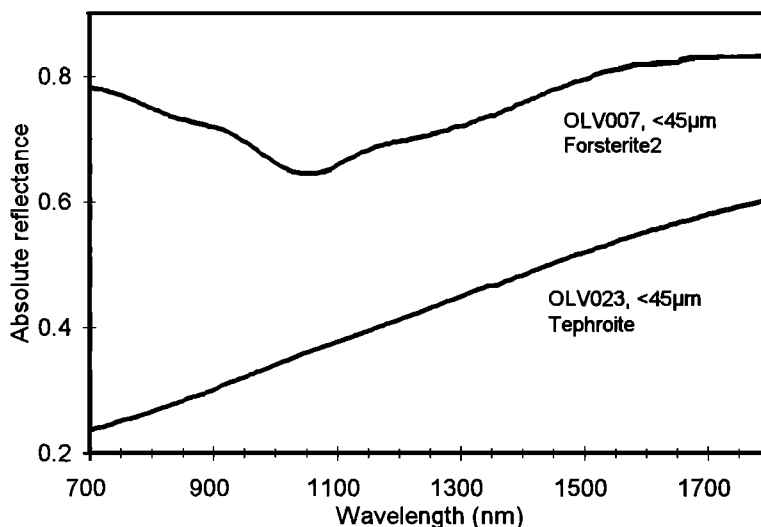


Figure 3. Reflectance spectra of the major ferrous iron absorption band region of an Mn-rich tephroite (OLV023;  $\text{Mg}_{8.3}\text{Fe}_{3.0}\text{Mn}_{78.2}$ ) and Mn-poor olivine (Forsterite2: OLV007;  $\text{Mg}_{95.9}\text{Fe}_{3.1}\text{Mn}_{0.2}$ ). See Table 1 for complete compositional data.

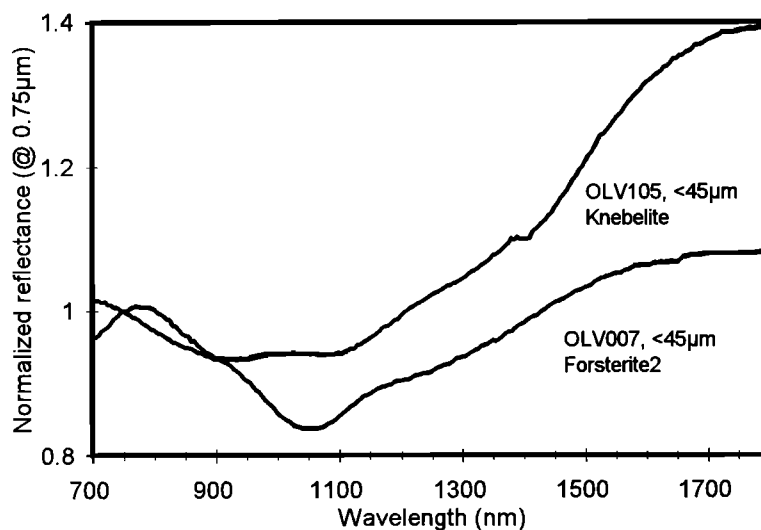


Figure 4. Normalized reflectance spectra (to 1 at 0.75  $\mu\text{m}$ ) of the major ferrous iron absorption bands region of an Mn-rich knebelite (OLV105;  $\text{Mg}_{3.4}\text{Fe}_{66.4}\text{Mn}_{29.7}$ ) and Mn-poor olivine (Forsterite2: OLV007;  $\text{Mg}_{95.9}\text{Fe}_{3.1}\text{Mn}_{0.2}$ ). See Table 1 for complete compositional data.

<45  $\mu\text{m}$  and 45-90  $\mu\text{m}$  grain sizes in the wavelength region of interest, but the narrow bands located near 0.400, 0.450, and 0.490  $\mu\text{m}$  are much more apparent in the larger fraction spectrum. This suggests that the absence of these bands could be attributed to more than one mechanism: a fine-grained forsterite-fayalite or an Mn-rich olivine; hence it is not a reliable method for discriminating Mn-rich from Mn-poor olivines.

### 3.4. $\text{Mn}^{2+}$ Spin Forbidden Bands

Burns [1970] also attributed a number of additional peaks seen in transmission spectra of fayalites-tephroites at 0.4095-0.4130  $\mu\text{m}$ , 0.4375-0.4400  $\mu\text{m}$ , and 0.5820-0.6000  $\mu\text{m}$  to  $\text{Mn}^{2+}$  transitions, the most prominent being the shortest wavelength band. Visible region reflectance spectra of the 45-90  $\mu\text{m}$  size fractions of the tephroite and knebelite are shown in Figure 6. Both spectra exhibit a readily detectable absorption feature near 0.41  $\mu\text{m}$ . While the spectral resolution (5 nm) is inadequate for detailed analysis of this feature, it seems to occur at a slightly shorter wavelength in the knebelite spectrum. This is consistent with the results of Burns [1970], who found that this feature shifted to longer wavelengths and became more intense as manganese content increased in transmission spectra.

The wavelength position of this band in the reflectance spectra is also within the range found by Burns [1970]. The additional

longer wavelength bands that were found by Burns in the transmission spectra, and which he attributed to  $\text{Mn}^{2+}$  transitions, are weaker than the band near 0.4  $\mu\text{m}$  in transmission and are not readily apparent in the reflectance spectra.

## 4. Discussion

Manganese preferentially occupies the M2 crystallographic site [Francis and Ribbe, 1980; Miller and Ribbe, 1985]. This has the effect of excluding  $\text{Fe}^{2+}$  from this site and results in a decrease in  $\text{Fe}^{2+}$  M2 absorption band strength. A decrease in olivine absorption band strength can be mimicked by a number of other effects such as particle size diminution [King and Ridley, 1987] or the presence of opaque minerals [Cloutis et al., 1991] and hence is not the most reliable indicator of the presence of Mn. However the decrease in  $\text{Fe}^{2+}$  M2 absorption band strength relative to the  $\text{Fe}^{2+}$  M1 absorption bands should be a useful indicator of the presence of appreciable Mn. This effect is apparent in the knebelite spectrum but a quantitative assessment of relative band strengths must await the acquisition and characterization of additional reflectance spectra and application of curve fitting algorithms such as the Modified Gaussian Method [Sunshine and Pieters, 1990]. Burns [1970] found a systematic change in relative band strengths in the transmission spectra of Mn-bearing olivines.

Table 2. Assignment of Spin-Forbidden  $\text{Fe}^{2+}$  Transition Bands by Various Investigators

Wavelength, $\mu\text{m}$	Reference
0.452-0.455; 0.495-0.500; 0.630-0.635	Burns [1970]
0.402, 0.425, 0.456, 0.472, 0.490, 0.554, 0.620	Burns et al. [1972]
0.630	White and Keester [1966]
0.450; 0.595	Vaughan and Burns [1977]
0.444; 0.508	Rager et al. [1988]
0.625	Hazen et al. [1977]
Numerous bands: 0.353-0.5	Grum-Grzhimailo et al. [1969]
Numerous bands: 0.35-0.65	Farrell and Newnham [1965]

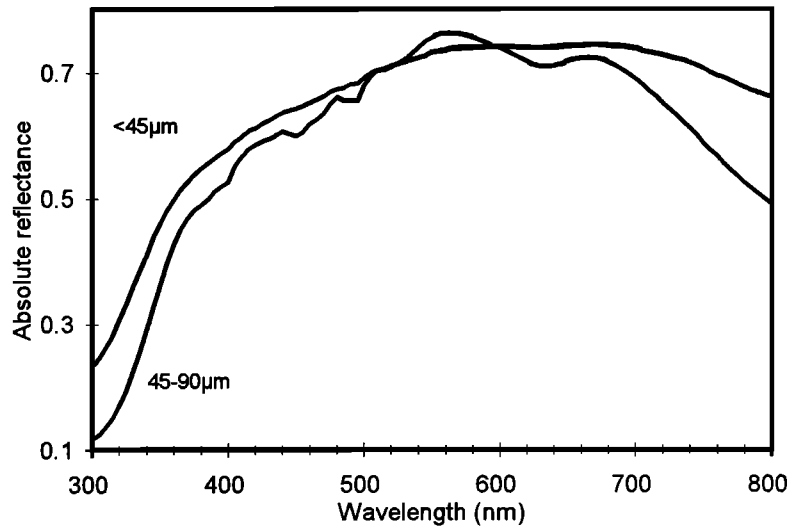


Figure 5. Absolute reflectance spectra of the visible wavelength region for two grain sizes (<45 and 45-90  $\mu\text{m}$ ) of an Mn-poor olivine (Forsterite1: OLV005;  $\text{Mg}_{89.8}\text{Fe}_{9.7}\text{Mn}_{0.1}$ ). See Table 1 for complete compositional data.

Determining manganese content from its effect on the wavelength position of the major  $\text{Fe}^{2+}$  absorption band near 1.05  $\mu\text{m}$  is a more reliable and more easily applied calibration. The available data, both from this study and transmission spectra [Burns, 1970], indicate that  $\text{Mn}_2\text{SiO}_4$  contents ranging from ~10 to ~70% result in a shift in the wavelength position of the  $\text{Fe}^{2+}$  M2 absorption band to longer wavelengths, outside the range expected for forsteritic-fayalitic olivines.

Such shifts would be relatively unaffected by grain size changes [Adams and Filice, 1967; King and Ridley, 1987] or the presence of spectrally neutral opaques [Cloutis *et al.*, 1991]. However, the presence of other minerals such as pyroxenes which also exhibit absorption bands in this wavelength region could shift the position of this band to longer or shorter wavelengths. The presence of these phases could be determined by other criteria, however [Cloutis and Gaffey, 1991a,b].

The absence of  $\text{Fe}^{2+}$  spin forbidden bands has been suggested as a possible indicator of manganese-rich olivines [King and Ridley, 1987]. While these bands are in fact absent in the reflectance spectra of knebelites and tephroites, they are often also absent in the reflectance spectra of fine-grained (<60  $\mu\text{m}$ ) Mn-poor olivines. Thus their presence or absence is not a reliable indicator of olivine composition.

Burns [1970] ascribed a number of visible region absorption features in transmission spectra of Mn-rich olivines to  $\text{Mn}^{2+}$  transitions. The most intense of these occurs in the 0.40-0.41  $\mu\text{m}$  region, and this feature is evident in the reflectance spectra of Mn-rich olivines. A similar feature is seen in the reflectance spectra of Mn-poor olivines, such as Forsterite1, and has also been attributed to  $\text{Fe}^{2+}$  spin forbidden transitions [Burns *et al.*, 1972]. The Mn-rich sample spectra exhibit only this feature in the visible region and not the additional ferrous iron bands. Thus the

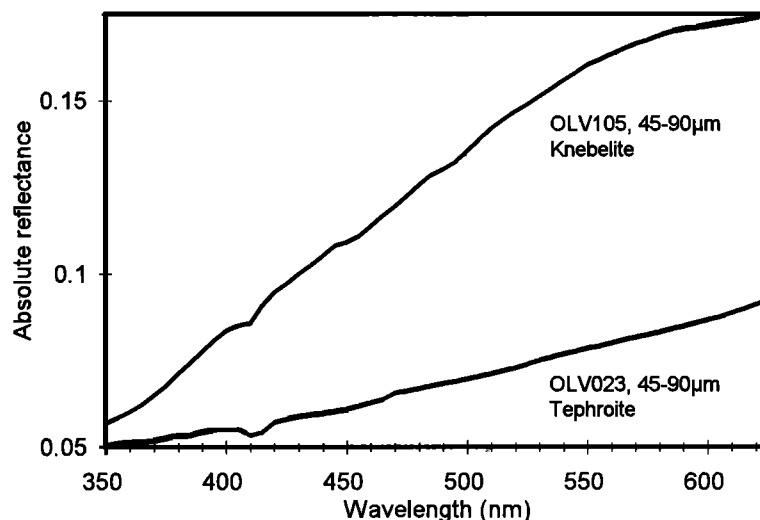


Figure 6. Absolute reflectance spectra of the visible wavelength region of 45-90  $\mu\text{m}$  grain size fractions of Mn-rich olivines: tephroite (OLV023;  $\text{Mg}_{8.3}\text{Fe}_{3.0}\text{Mn}_{78.2}$ ) and knebelite (OLV105;  $\text{Mg}_{3.4}\text{Fe}_{66.4}\text{Mn}_{29.7}$ ). See Table 1 for complete compositional data.

presence of only this feature in the visible region can be used to distinguish Mn-rich from Mn-poor olivines. If additional absorption features, attributable to ferrous iron, are present in the visible region, the use of this band to identify Mn-rich olivines is not justified.

## 5. Conclusions

When a number of spectral features are taken together, they can be used to distinguish manganese-rich from manganese-poor olivines from reflectance spectra. Mn-rich olivine spectra will be of two types. Those which do not exhibit ferrous iron absorption features in the 1  $\mu\text{m}$  region will be the most Mn-rich (> 70%  $\text{Mn}_2\text{SiO}_4$ ) and will exhibit a single  $\text{Mn}^{2+}$  absorption feature in the 0.40-0.41  $\mu\text{m}$  region. Those which do exhibit a ferrous iron absorption feature in the 1  $\mu\text{m}$  region will have more prominent  $\text{Fe}^{2+}$  M1 absorptions relative to  $\text{Fe}^{2+}$  absorptions than Mn-poor olivines, the minimum reflectance will occur at wavelengths outside the range of forsterite-fayalite (greater than ~1.08  $\mu\text{m}$ ), and they will exhibit a single prominent  $\text{Mn}^{2+}$  transition band in the 0.40-0.41  $\mu\text{m}$  region.

These results indicate that manganese-rich olivines are spectrally distinct from forsteritic-fayalitic olivines and that visible and near-infrared reflectance spectra can be used to detect their presence on planetary surfaces.

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