

Water on Venus: New Insights from Tremolite Decomposition

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Venus is an extremely hot and dry planet with very little water in its atmosphere and no standing water on its surface. However, if Venus had had more water in the past, as the high D/H ratio implies, hydrous minerals might have formed. We present the first experimental study that examines the decomposition rate of hydrous minerals, specifically tremolite, with applications to Venus. We predict that tremolite can survive decomposition on Venus over geologic time scales at current and at higher surface temperatures. Thus, if future spacecraft can detect tremolite, this discovery could serve as evidence of a wetter Venusian history. © 2000 Academic Press

Key Words: Venus; Venus, surface; tremolite; water; mineralogy.

INTRODUCTION

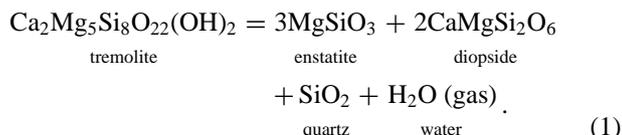
Venus is an extremely hot and dry planet where surface temperatures range from 660 to 740 K and water vapor in the subcloud atmosphere is present at ~30 parts per million by volume (ppmv). In contrast, Earth's average surface temperature is 288 K and its troposphere is much wetter with a water vapor content up to ~4% (Lodders and Fegley 1998). Even though these planets formed next to each other within the inner Solar System, they have very different characteristics. We might reasonably expect that both planets would have accumulated similar amounts of water during accretion. However, the extreme differences between the two planets lead us to the long-standing question: has Venus always been this dry, or did it lose its water over time?

The first clue that suggests that Venus may have had more water in its past is the high deuterium/hydrogen (D/H) ratio of $(2.5 \pm 0.5) \times 10^{-2}$ in atmospheric water vapor (Donahue *et al.* 1997). This is 160 ± 32 times the D/H ratio of 1.56×10^{-4} in standard mean ocean water (SMOW) on Earth (Lodders and Fegley 1998). During isotope fractionation, hydrogen preferentially escapes through physical processes such as evaporation or diffusion, thus enriching the deuterium concentration (e.g., see Criss 1999). The D/H ratio of Venus's atmosphere was studied by Donahue *et al.* (1982, 1997, and references therein). They concluded that Venus once must have had the equivalent of at least a 4-m-deep global ocean and may have had as much as a 530-m-deep global ocean. However, some interplanetary dust particles, meteorites, and comets also have high D/H ratios (Irvine *et al.*

2000). Therefore, it is possible that Venus could have accreted matter with different D/H ratios than did Earth. Even though the high D/H ratio is a good indicator, by itself, it is only indirect proof that Venus was once wet.

A second clue to the history of water on Venus could be hidden within the surface mineralogy. If there had been more water in Venus' past, it is possible that hydrous minerals formed on or within the surface. However, equilibrium thermodynamic calculations predict that these minerals are unstable at venusian surface temperatures and pressures (Zolotov *et al.* 1997). Notwithstanding, the planetary community has debated the existence of hydrous minerals on Venus for over 30 years without knowledge of the rate at which hydrous minerals decompose (e.g., Mueller 1964, Lewis 1970, Khodakovsky 1982). This is the first study that investigates hydrous mineral decomposition rates with applications to Venus. Preliminary results are reported by Johnson and Fegley (1998, 2000).

Hydrous minerals, such as amphiboles, are fairly common on Earth in metamorphic regimes and have also been detected in limited amounts in both SNC meteorites and lunar samples (Fronzel 1975, Deer *et al.* 1997, McSween and Treiman 1998). We chose to study tremolite, a relatively common calcic end-member amphibole, because (1) it has a simple composition, (2) its structure is well known, (3) relatively pure samples are easy to obtain, and (4) previous work by other authors shows what decomposition products to expect (e.g., Posnjak and Bowen 1931, Boyd 1959, Freeman and Taylor 1960, Xu *et al.* 1996). At Venus' modal radius of 6051.4 km (Ford and Pettengill 1992), total pressure is 95.6 bar (Seiff *et al.* 1986) and water vapor is present at 30 ppmv (e.g., see de Bergh *et al.* (1995) and the H₂O measurements tabulated in Fegley *et al.* (1997)). This gives a water partial pressure of ~3 millibar at the surface of Venus. In order for tremolite to be currently stable on the plains of Venus at 740 K, the atmospheric partial pressure of water vapor must be at least 6 bar to prevent tremolite decomposition via the following reaction (Zolotov *et al.* 1997):



This value is approximately 2000 times greater than the current H_2O partial pressure on Venus; therefore tremolite should decompose. However, the existence of tremolite on Venus cannot be summarily dismissed because the rate of reaction (1) is unknown. For example, diamond is thermodynamically unstable at Earth's surface, yet we do not observe diamonds turning into lumps of graphite. If tremolite formed on or (more likely) below the surface during a wetter era in Venus' past, our results suggest that there is a good possibility that tremolite could presently exist on Venus.

TREMOLITE DECOMPOSITION EXPERIMENTS

We studied the kinetics of reaction (1) by isothermally heating tremolite powder samples of known weight, surface area and composition in dry CO_2 or N_2 gas. Experiments were run with tremolite from Canaan, CT supplied by Ward's Natural Science. Tremolite grains were hand picked, ground into powder, and then agitated in HCl to remove possible carbonate contamination. Electron microprobe and X-ray diffraction (XRD) results show that the treated powder was pure tremolite. Samples were run in either instrument grade CO_2 (99.99%) or ultrahigh purity (UHP) N_2 (99.999%). Carbon dioxide (96.5%) and N_2 (3.5%) are the two most abundant gases in Venus' atmosphere (Lodders and Fegley 1998). The gases used in the experiments typically contain 10 ppmv water vapor which is similar to the subcloud atmospheric concentration on Venus. Prior to and after heating, each sample was weighed to measure water loss. About 200 experiments, ranging in duration from hours to over 7 months, were done at temperatures of 1045–1238 K. These experiments were run at higher temperatures than those at Venus' surface so that decomposition would occur in a reasonable length of time. The results show that tremolite decomposes at the same rate in both gases.

When tremolite is heated and decomposes, it loses water. We determined the extent of reaction as a function of time by measuring the weight (water) loss. A typical weight loss curve is shown in Fig. 1. Similar curves were observed by Clark and Freeman (1967) who studied the thermal decomposition of crocidolite, an Fe-bearing amphibole known as blue asbestos. Figure 2 shows the fraction of tremolite reacted plotted versus time normalized to the time required for 50% decomposition of the tremolite (i.e., $t/t_{0.5}$). This type of plot (e.g., see Bamford and Tipper 1980) shows that tremolite decomposition follows the same kinetic behavior and occurs by the same mechanism (although at a different rate) at all temperatures we studied. Figure 2 also shows that complete decomposition of tremolite takes several to 10 times longer than the time required for 50% decomposition.

Decomposition of tremolite was confirmed by electron microprobe analyses, XRD, and mid-infrared spectroscopy. We used the Washington University JEOL-733 electron microprobe (15 kV accelerating voltage, 30 nA beam current), Rigaku vertical powder diffractometer with $\text{CuK}\alpha$ radiation, and a Fourier transform infrared Bomen DA3.02 spectrometer with HgCdTe

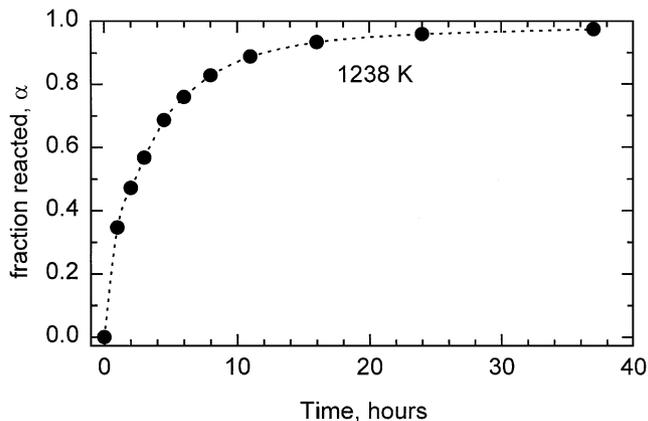


FIG. 1. A typical weight loss curve for tremolite decomposition, in this case at 1238 K. The fraction reacted is plotted as a function of time. The fraction reacted is calculated from the weight loss. When half of the tremolite has decomposed, the observed weight loss is half of the complete water (weight) loss and the fraction reacted is 0.5. When all tremolite has decomposed, the observed weight loss reaches its maximum and the fraction reacted is one.

detector. Figure 3 shows the decrease of the mid-infrared hydroxyl (OH) peak (3674 cm^{-1}) versus extent of decomposition. The infrared and electron microprobe data demonstrate that the observed weight loss is due to water loss and not to another reaction.

KINETIC RESULTS

The reacted samples were studied by scanning electron microscopy (SEM) using the McDonnell Center for Space Sciences JEOL JSM-840A scanning electron microscope (5 kV accelerating voltage, 50 pA beam current). The SEM images show that decomposition proceeds as a combination of nucleation and growth of reacted regions. Diffusion of water through the reacted layers is not a controlling factor because the reacted areas are

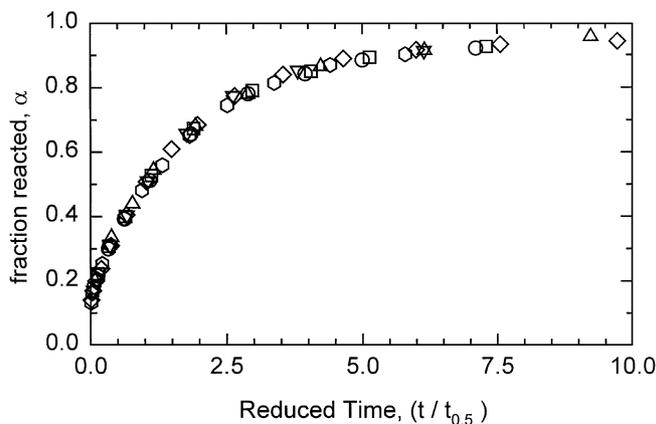


FIG. 2. A plot of the fraction of tremolite reacted plotted versus time normalized to the 50% decomposition time ($t_{0.5}$). The different symbols are for tremolite heated at different temperatures in nitrogen: Δ , 1237 K; \square , 1213 K; \circ , 1202 K; ∇ , 1177 K; \diamond , 1150 K; \circ , 1122 K.

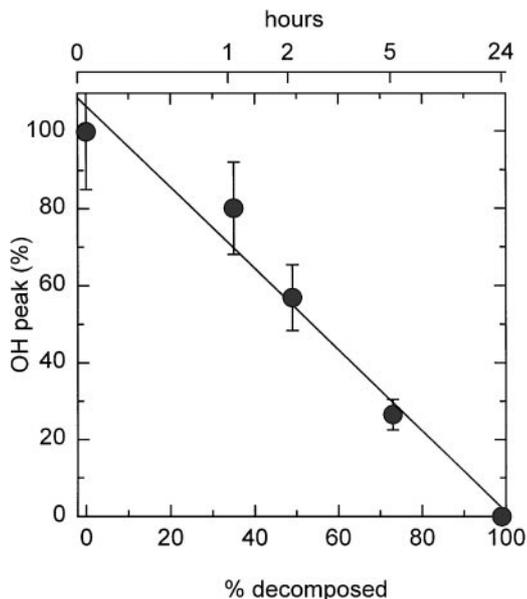


FIG. 3. This plot shows the decrease of the tremolite hydroxyl (OH) peak (3674 cm^{-1}) with extent of decomposition. The peak was measured by mid-IR spectroscopy. The error bars are $\pm 15\%$ and caused by variation in the sample thickness.

extremely porous. The images in Fig. 4 illustrate a crystalline tremolite sample prior to and after heating at 1224 K. Decomposition appears to occur as random nucleation on the edges and at crystal defects (e.g., cracks) with the reaction following the original crystal lattice. As these decomposed regions expand and envelope previously reacted areas, new nucleation sites become visible within the unaffected areas. Figure 4 shows that tremolite thermal decomposition is very different than pyrite thermal decomposition where the reaction interface is well defined and advances inward linearly with time (e.g., see Fig. 4 of Fegley *et al.* (1995) and Fig. 3 of Hong and Fegley (1997)).

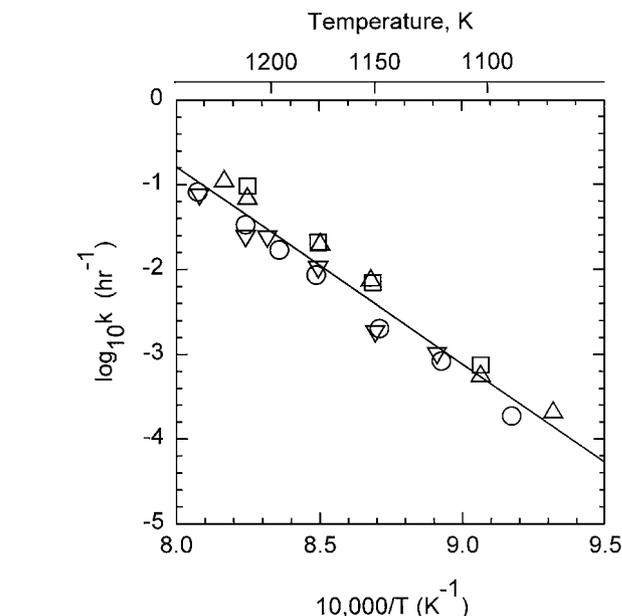
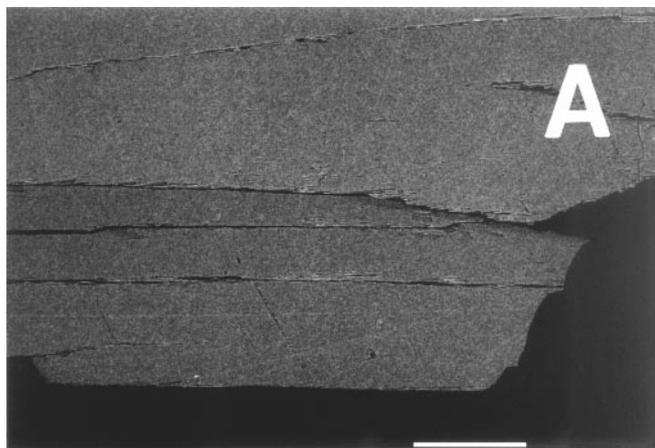


FIG. 5. Reaction rate constants versus $1/T$ shown in an Arrhenius plot. \circ , vertical furnace, CO_2 gas, beaker style crucible; ∇ , vertical furnace, N_2 gas, beaker style crucible; \square , horizontal furnace, CO_2 gas, beaker style crucible; \triangle , horizontal furnace, CO_2 gas, boat style crucible. One sigma error bars are the size of the symbols on the plot.

The Avrami-Erofe'ev rate model best describes what we observe in the SEM images. The rate equation for the Avrami-Erofe'ev model is $kt = (-\ln(1 - \alpha))^{1/2}$, where k is the rate constant (h^{-1}), t is time, and α is the fraction reacted of the sample ($\alpha = \% \text{ wt. loss} / \% \text{ theoretical wt. loss}$, which is 2.22%). This model takes into account random nucleation and growth given that as the nuclei grow, nucleation sites are both generated and removed (Bamford and Tipper 1980).

The Arrhenius plot in Fig. 5 shows the linear relationship between the log of reaction rate constants and inverse temperature.

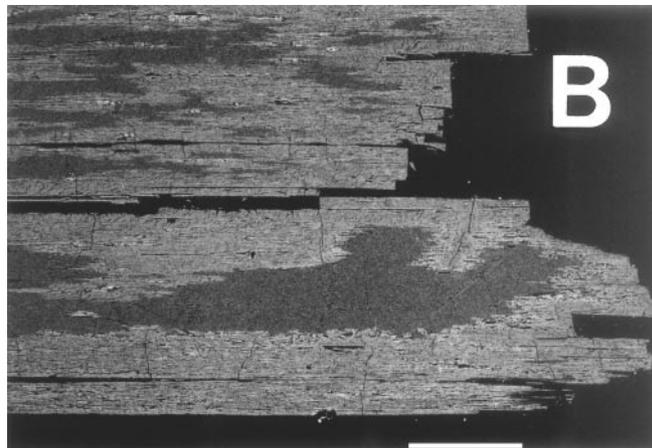


FIG. 4. Scanning electron micrographs of (A) unreacted tremolite and (B) 60% decomposed tremolite after heating at 1224 K. Decomposed regions are lighter in hue and porous. Unreacted material in B is darker and located within the center of the grain. Scale bar is $100\ \mu\text{m}$ for both images.

TABLE I
Rate Data Used to Calculate
the Activation Energy

Temp (K)	Rate $\times 10^{-3}$ (h $^{-1}$) ^b
N ₂ gas ^a	
1237	76 \pm 7
1213	25 \pm 2
1202	24 \pm 2
1177	11 \pm 1
1150	1.9 \pm 0.2
1122	1.0 \pm 0.1
CO ₂ gas set 1 ^a	
1238	82 \pm 7
1213	33 \pm 3
1196	17 \pm 1
1178	8.6 \pm 0.6
1148	2.0 \pm 0.2
1120	0.83 \pm 0.06
1090	0.19
CO ₂ gas set 2 ^c	
1212	96 \pm 8
1177	21 \pm 2
1151	7.0 \pm 0.4
1103	0.75 \pm 0.06
CO ₂ gas set 3 ^d	
1224	110 \pm 5
1212	68 \pm 8
1176	20 \pm 2
1152	7.4 \pm 0.9
1103	0.56 \pm 0.11
1073	0.21 \pm 0.01

^a Vertical furnace, beaker style crucible.

^b Errors are 1 σ .

^c Horizontal furnace, beaker style crucible.

^d Horizontal furnace, boat style crucible.

The Arrhenius equation is $k = A \exp(-E_a/RT)$, where A represents the frequency factor (collision frequency), T is the temperature in Kelvin, R is the gas constant, E_a is the activation energy, and k is the rate constant. The mean rate constant for each temperature is given in Table I. Analysis of the data gives the rate equation

$$\log_{10} k \text{ (h}^{-1}\text{)} = 17.85(\pm 0.21) - 23,299(\pm 1215)/T, \quad (2)$$

where T is in Kelvin with a corresponding activation energy of $446 \pm 23 \text{ kJ mol}^{-1}$.

DISCUSSION

(a) *Tremolite survival on Venus.* The rate of reaction (1) was extrapolated to Venus surface temperatures (~ 660 – 740 K) using the derived rate Eq. (2). Table II gives the temperatures and estimated times for 50% decomposition of powdered tremolite grains as used in these experiments. These grains have a specific

surface area of $4.86(\pm 0.42 \text{ at } 1\sigma) \text{ m}^2 \text{ g}^{-1}$ as measured by N₂ gas adsorption with a Quantasorb surface area analyzer. The measured surface area corresponds to an equivalent spherical diameter of $0.40 \mu\text{m}$. Table II shows that it becomes increasingly difficult for tremolite to decompose as the temperature decreases. Thus, if a $0.40\text{-}\mu\text{m}$ -diameter grain of powdered tremolite were placed on Venus' lowlands at 740 K today, the grain would reach 50% decomposition in 3.8 billion years. Furthermore, the normalized time plot in Fig. 2 shows that complete decomposition of tremolite at 740 K would take significantly longer. One tremolite sample has been heated at 740 K for 21 months and has not decomposed. Further heating of this sample is ongoing, but we do not expect any decomposition to occur. Our findings from all our experiments suggest that tremolite grains do not decompose in geologic time scales at current venusian surface conditions.

However, computer models that are currently under debate, suggest that Venus might have been 100 K hotter in the past than today (Bullock and Grinspoon 1999). This high-temperature excursion may have persisted for 100 to 400 Ma. Even so, the chances of tremolite survival are still good. In the venusian highlands, at the 100° higher temperature (760 K), $0.40\text{-}\mu\text{m}$ -diameter tremolite grains would reach 50% decomposition in 500 Ma, which is longer than the duration of the higher temperatures. Complete decomposition would take ~ 5.0 billion years (see Fig. 2). Larger grains would take even longer to decompose.

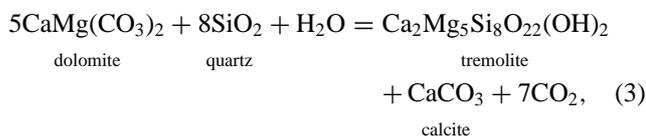
Chemical weathering on the surface could also affect tremolite survival. Sulfur dioxide is the third most abundant gas in Venus' subcloud atmosphere with a mixing ratio of 150 ppmv (Lodders and Fegley 1998). Zolotov *et al.* (1997) calculated that at equilibrium, tremolite will be attacked by SO₂ to form anhydrite (CaSO₄), enstatite, quartz, and water vapor. We checked the importance of this postulated reaction by heating tremolite in a 1% SO₂–99% CO₂ gas mixture at 1106 K for 13 days. The XRD patterns are the same as those observed when tremolite is heated in CO₂ under similar conditions. There is no evidence for anhydrite formation, which occurs within a few days when calcite (CaCO₃) or diopside are heated at about the same temperature in a 1% SO₂–99% CO₂ gas mixture (Fegley and Prinn 1989a). Thus, our preliminary experiments at high temperature indicate that tremolite thermal decomposition occurs faster than tremolite reaction with SO₂ to form anhydrite.

TABLE II
Time for 50% Decomposition
(0.4 μm grains)

T (K)	Time
1200	31 h
1100	74 Days
1000	27 Years
900	10,000 Years
800	18 Ma
740	4 Ga

(b) *Tremolite formation.* Yet, the question remains, how much tremolite could have formed on Venus in the past? This depends on how much water was available. We can constrain the tremolite abundance from an estimate of the abundance of water on Venus in the past. If Venus once had $\sim 1100 \mu\text{g}$ of water per gram of rock, which is an estimate for the bulk silicate Earth (Zolotov *et al.* 1997 and references therein), and all this water were in tremolite, then tremolite would exist at ~ 5.0 mass%. Although this is probably an upper limit, percentage levels of tremolite are plausible. If we refer to terrestrial geology, metamorphic rocks typically contain a few percent amphiboles.

It is reasonable to postulate that hydrous minerals could have formed on a wetter Venus. On Earth, we find tremolite in low-grade metamorphic ultrabasic rocks and in contact metamorphosed siliceous carbonate rocks (Deer *et al.* 1997). Geochemical arguments suggest the presence of both rock types on Venus (Fegley *et al.* 1992, Kargel *et al.* 1994, Fegley *et al.* 1997). One possible reaction for forming tremolite is



which occurs on Earth (Deer *et al.* 1997), but other reactions are also possible. Tremolite also occurs as an alteration product of pyroxenes that interact with water-enriched magmatic fluids (Deer *et al.* 1997). Under sufficiently high water partial pressure, it is possible that tremolite would have formed in localized areas of Venus' crust via magmatic heating. Zolotov *et al.* (1999) also discuss potential locations for hydrous mineral formation on Venus. If we accept that there was a significant water component on Venus in the past, then it would likely be present in the right settings to form hydrous minerals similar to those on Earth.

Likely regions that might favor the survival of amphiboles are the highlands, tesserae, and/or crater ejecta blankets. The mineralogy of the tesserae, which cover $\sim 8\%$ of Venus' surface (Ivanov and Head 1996), is unknown. If the tesserae are metamorphic, as proposed by Nikolaeva *et al.* (1992), they may be a good locale for preserving tremolite-bearing rocks. The highlands and tesserae are at higher elevations that have cooler temperatures which favor tremolite preservation. Last, crater ejecta blankets could contain amphiboles from older excavated layers that were brought to the surface upon impact.

(c) *Mineral detection.* Unfortunately, no direct mineralogical data exist for the venusian surface. The only geochemical data we have for the surface are abundances of Mg and heavier elements measured by XRF spectrometers on the Venera 13 and 14 and Vega 2 landers (Surkov *et al.* 1984, 1986). Unlike the Moon and Mars, there are no known meteorite samples from the venusian surface. What information we have about the surface mineralogy was derived through thermodynamic modeling

and deductions based on the XRF elemental analyses and orbital radar (Pettengill *et al.* 1982, Barsukov 1992, Fegley and Treiman 1992, Fegley *et al.* 1992, Kargel *et al.* 1994, Brackett *et al.* 1995, Fegley *et al.* 1997).

The best method to search for tremolite and other amphiboles is through direct mineralogical measurements via landers, penetrators, rovers, and/or automated balloons (aerobots). Indirect methods such as *in situ* neutron spectroscopy or thermogravimetric/differential thermal analysis (TGA/DTA) can be used to detect hydrous minerals, but these techniques are unlikely to be useful on Venus (Zolotov *et al.* 1997). Preferred techniques that can identify hydrous minerals on Venus include XRD and/or imaging infrared reflectance spectroscopy (Blake *et al.* 1994, Rossman 1988). Imaging infrared reflectance spectroscopy can be used to distinguish between chemically bound water in minerals and other forms of water, and to identify specific hydrous minerals. It seems to be the method of choice for detecting hydrous minerals on Venus, although development and testing of a suitable spacecraft instrument is necessary (e.g., Zolotov *et al.* 1997). Potential landing sites that might retain amphiboles include crater ejecta blankets and areas at higher and cooler elevations, such as the highland plateaus and tesserae. It is important to be mobile and analyze several different locations because a single sample would be of limited value.

SUMMARY

If we are able to detect amphiboles or other thermally stable hydrous minerals (Zolotov *et al.* 1997) on the surface, we will know that Venus once had water that participated in geologic, geochemical, and atmospheric processes. Such a discovery would answer the long-standing question posed in the introduction with the response: Venus lost its water over time. Theory tells us that the formation of hydrous minerals was not favored within the inner portion of the solar nebula because of kinetic limitations and that hydrous minerals would not have been available during accretion (Fegley and Prinn 1989b, Fegley 2000). Consequently, this lack of hydrous minerals in the inner solar nebula suggests that water was delivered to Venus, Earth, and Mars from beyond the inner Solar System via icy bodies and/or hydrous mineral bearing planetesimals. Knowing that tremolite can survive decomposition on Venus over geologic time scales, it is important to send spacecraft that can detect hydrous minerals because such a discovery would provide ground truth confirming that Venus had more water in its past.

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