#### LETTER

# Evidence in favor of small amounts of ephemeral and transient water during alteration at Meridiani Planum, Mars

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### ABSTRACT

In light of the controversy surrounding the origin of sulfate-rich rocks analyzed by the Mars Exploration Rover Opportunity, thermodynamic and kinetic models explored the consequences of in situ alteration of basaltic sand by pristine sulfuric acid. Simulations at 273 K and current martian atmosphere show that it is possible to simultaneously account for both chemical and mineralogical observations at the Meridiani landing site, but only when the amounts of water are small (water/rock mass ratio  $\leq 1$ ), the aqueous solutions are highly acidic (pH < 3), and the lifetimes of liquid water are extremely short (on the order of tens of years). Furthermore, the best agreement between observations and models is obtained if evolved fluids are removed after alteration. If this simple self-consistent scenario is relevant to bedrock formation at Meridiani, it provides stringent constraints on the issues of where and when liquid water was present at the surface of Mars.

Keywords: Mars, alteration, Meridiani, thermodynamic modeling, sulfuric acid, SO<sub>3</sub>

#### INTRODUCTION

Constraining where and when liquid water was present at the surface of Mars has been a constant motivation for exploration of the red planet. In this respect, one of the most promising ways to highlight the presence of water on Mars in the geological past is to look for secondary minerals characteristic of the interaction of pristine magmatic rocks with aqueous solutions. Thanks to the instruments onboard recent landed and orbital missions, an unprecedented amount of high-quality data exists relevant to this question. In this respect the sulfate-rich bedrocks analyzed by the rover Opportunity in Eagle and Endurance craters on Meridiani Planum (Squyres et al. 2004; Grotzinger et al. 2005; Clark et al. 2005; McLennan et al. 2005) provide important constraints on the physical and thermodynamic conditions at the time of sulfate deposition. Although laboratory and theoretical studies demonstrate that acidic aqueous solutions played an important role (Chevrier and Mathé 2007; Hurowitz et al. 2006), significant controversy surrounds many critical aspects of Meridiani bedrock formation (McCollom et al. 2005, 2006; Squyres et al. 2006). In the model developed by the Mars Exploration Rover (MER) team (McLennan et al. 2005; Squyres et al. 2004, 2006), the stratigraphic and geochemical sequences observed are explained by deposition (dominantly aeolian) of previously altered sand grains, into which low-temperature brines rich in sulfate salts are introduced during open-system diagenetic events linked to variations in the local water table. This view has been challenged by McCollom and Hynek (2005) who inferred local hydrothermal interaction of unaltered volcanic sands with high-temperature, sulfur-rich aqueous solutions. This scenario was motivated by the

fact that no obvious large sedimentary basin capable of producing large quantities of altered sand grains is apparent, in addition to the observation that, to a first approximation, the composition of Meridiani bedrock is consistent with simple addition of sulfur to an isochemically altered basalt. The importance of this latter observation was refuted by Squyres et al. (2006) who stressed the existence of geochemical trends to a (Mg,Ca,Fe)SO<sub>4</sub> component, taken as evidence of open-system behavior.

Although models involving in situ alteration of basaltic sands have the advantage that they alleviate the need for large unidentified external sources of altered grains and/or brines, preliminary thermodynamic simulations performed assuming a closed system did not reproduce the mineralogy observed at the Opportunity landing site (McCollom and Hynek 2005; Tréguier et al. 2008). Furthermore, only limited attempts have been made to assess whether the geochemical trends highlighted by Squyres et al. (2006) are consistent with the scenario of in situ alteration, although McCollom and Hynek (2005) briefly addressed this question by envisaging local migration of magnesium sulfate salts within the sediments. Tréguier et al. (2008) considered the data set obtained by the alpha particle X-ray spectrometer (APXS) onboard the MER rovers, and used principal component analysis to highlight the addition of a pure sulfur component in rocks at Meridiani. These authors also used thermodynamic models to quantify the alteration of martian basalt by sulfuric acid in a closed system, considering bulk dissolution of the pristine basalt, or preferential leaching of pyroxene and olivine. These closed-system calculations suggested that the observed mineralogy requires much larger additions of sulfur than the value deduced from rock analyses.

With these issues in mind, we have extended the approach of Tréguier et al. (2008), taking into account more realistic kinetic constraints to model basalt alteration as a function of

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time, with the aim of identifying self-consistent scenarios for rock-formation at Meridiani.

## **EXPERIMENTAL METHODS**

The thermodynamic calculations were performed using the geochemical simulator JChess (Van der Lee and De Windt 2002), a numeric tool equivalent to EQ3/6, PhreeqC, or Geochemist's Workbench, using the SUPCRT92 thermodynamic database (Johnson et al. 1992). JChess simulates the mass transfer between gas, solids, and an aqueous solution during irreversible reaction and the calculation proceeds by a succession of increments, in which the thermodynamic equations describing solid-aqueous solution equilibria are solved at each time step. The nature and amount of secondary phases and coexisting aqueous solution composition are calculated as a function of time. The activities of aqueous species were calculated by B-dot equations rather than the Pitzer formalism. Although the latter would be more appropriate at high ionic strength, insufficient data are available to constrain models for the multi-component chemical system considered here. No modification of bulk composition was introduced between time steps, but the geochemical consequences of brine mobility on the composition of the altered rock sub-system may nevertheless be quantified by considering addition or removal of the brine component.

## **RESULTS AND DISCUSSION**

The initial silicate material modeled is unaltered olivinerich martian basalt Adirondack described from Gusev crater (McSween et al. 2006). The initial fluid phase is assumed to be pristine sulfuric acid, derived from the oxidation and dissolution of volcanically produced (or possibly impact-generated) sour gas in pure water. In this respect if SO<sub>2</sub> were the only oxidized sulfurbearing gas, the pH of the aqueous solution could not be much less than 4, and sulfites rather than sulfates would be produced by reaction with crystalline silicates (Halevy et al. 2007). On the other hand, SO<sub>3</sub> is highly soluble and capable of producing concentrated H<sub>2</sub>SO<sub>4</sub> solutions. Several formation mechanisms for SO<sub>3</sub> are possible (Settle 1979; Bullock and Moore 2007) and in this way significant amounts of sulfate can be produced during surface alteration, without the need for large volumes of water. Scenarios such as embedded dunes or aqueous films on surface particles (Niles and Michalski 2009) can therefore be envisaged. For the purpose of our calculations, alteration was simulated at 0 °C (ice melting) and partial pressures for O2 and CO2 equivalent to values observed in the current martian atmosphere were assumed. Tests show that results are not strongly dependent on temperature and that even variations of several orders of magnitude in the partial pressure of CO<sub>2</sub> have little effect under acidic conditions (the focus of this study) because for solutions of pH <4 carbonates do not precipitate, even under several hundred bars of CO<sub>2</sub> (Bullock and Moore 2007). Indeed, for the low water-rock ratio and low pH conditions considered here, rock alteration is above all driven by the number of protons, a quantity which may be characterized by the parameter "SO<sub>3</sub>" (corresponding to dry  $H_2SO_4$ ). Rock alteration will thus be a function of the mass ratio SO<sub>3</sub>/basalt, water simply acting as a medium allowing the chemical reactions to proceed and stabilizing hydrated minerals.

Initial calculations were performed with no kinetic constraints and assuming congruent dissolution (i.e., no primary phases remain in the altered part of the rock). These simulations are similar to those of Tréguier et al. (2008) but at lower water-rock ratio (W/R = 1). A range of SO<sub>3</sub>/basalt mass ratio from 0 to ~1 was modeled. If produced in one single event, the production of an altered basalt layer with SO<sub>3</sub>/basalt ~1 and a vertical dimen-

sion of 5 m (typical of the Burns formation in Endurance crater) would require scavenging of an atmospheric column 2 km high, for an initial pressure of 1 bar SO<sub>3</sub>. These values appear large, and production of enough sulfur to form regional deposits several hundred meters thick is an obvious problem. However, although the question of the source of sulfur is outside the scope of the present study, interesting insights are provided by the recent work of Gaillard and Scaillet (2009). Returning to the issue of alteration, hydrolysis of basalt through interaction with sour gas can be viewed as an exchange between the alkali and alkali-earth cations of the rock (and even aluminum and iron if pH is low enough), and the protons of the aqueous fluid. If conditions are sufficiently alkaline, addition of atmospheric O2 and CO2 may act to oxidize iron and precipitate carbonate, respectively. As a consequence, at low SO<sub>3</sub>/basalt the acidic solution is efficiently neutralized, producing a secondary mineral assemblage dominated by clays and carbonates (Fig. 1). Similar results have been calculated by McAdam et al. (2008a) for brines produced by basalt-H<sub>2</sub>SO<sub>4</sub> interactions. On the other hand, at the highest modeled SO<sub>3</sub>/basalt, alteration produces amorphous silica and direct precipitation of jarosite (rather than by evaporation of brine), the latter favored by the combination of low W/R ratio and extremely low pH. In this respect there are some similarities to results of Schiffman et al. (2006) concerning acid-fog alteration of Kilauea Volcano in Hawaii. Within the framework of our scenario the presence of jarosite in abraded Meridiani bedrock (rather than clays + gypsum) therefore implies  $SO_3$ /basalt > 0.5 if rock alteration had been total (Fig. 1). However, such values are considerably higher than bulk-rock sulfur contents measured by Opportunity which suggest SO<sub>3</sub>/basalt ratios on the order of 0.25 (Fig. 1).



FIGURE 1. Modeled alteration of bulk Adirondack basalt as a function of SO<sub>3</sub>/basalt mass ratio. For kinetic reasons the precipitation of quartz (replaced by chalcedony), hematite (replaced by goethite), and secondary mafic minerals is suppressed in the calculations. The lower panel presents secondary mineralogy as a function of SO<sub>3</sub>/basalt. The pie charts indicate the pH and compositions of the coexisting brines for the highest and lowest values of modeled SO<sub>3</sub>/basalt.

This discrepancy may be explained if the initial basaltic sand experienced incomplete alteration, with certain primary phases being more susceptible to attack than others. Indeed, the presence of pyroxene and/or plagioclase, but absence of olivine in the rocks of Eagle and Endurance craters (Morris et al. 2006; Glotch et al. 2006) provides strong independent evidence in favor of this hypothesis.

Additional kinetic simulations (i.e., as a function of time) have therefore been performed, using specific dissolution kinetics for each rock-forming mineral. Dissolution rates were calculated for initial grains of 1 mm, using dissolution constants derived from laboratory data at low pH (Zolotov and Mironenko 2007; McAdam et al. 2008b) and taking into account a first-order kinetic law for deviation from equilibrium and a 0.3 order pH dependence. The dissolution rate is modified at each step in the calculation as a function of changes in the composition and pH of the solution. Precipitation of chalcedony, goethite, carbonates, and sulfates is assumed to be instantaneous when saturation is reached, whereas the precipitation law used for clays is derived from feldspar dissolution kinetics (but of opposite sign). The time-scales of alteration calculated here are best estimates based upon current knowledge, but it should be appreciated that significant uncertainty surrounds the absolute values. For example, it has been shown that dissolution rate constants derived from experimental studies tend to overestimate the rate at which alteration occurs (e.g., Brantley 2003). Furthermore, the value of exposed mineral surface area has a significant effect on alteration rate, but values relevant to Meridiani are poorly constrained. These issues should therefore be borne in mind when considering derived time-scales. On the other hand, inaccuracies in the solubility data (e.g., absence of solid solutions and sparse data for nontronite in the Supert92 database) are of lesser importance, although they may have a minor influence on the value of pH at which the transition occurs from an alteration assemblage dominated by clays and carbonates to one dominated by sulfates and silica. A first set of calculations was performed to simulate closed system alteration along the lines proposed by McCollom and Hynek (2005). In this case, the bulk-rock sulfur content of Meridiani samples provides a direct estimate of the  $SO_3$ /basalt ratio to be used (~0.25). Of particular interest in this scenario is the question of whether it is possible to explain the compositional variations highlighted by Squyres et al. (2006) in terms of a variation in local proportions of altered solids and coexisting brine. Based upon a series of simulations with W/R from 0.5 to 100, we find that the Meridiani trend lies along such a mixing line (between altered solids and coexisting brine) only at  $W/R \le 1$  and for short reaction times, before neutralization of the fluid. However, when the calculated mineralogy of the solid fraction is considered as a function of time (Fig. 2b), it is found that at time-steps which are satisfactory from a chemical perspective (around 2 years at W/R = 0.5), significant quantities of unreacted olivine remain, in contrast to spectroscopic observations (Morris et al. 2006). We therefore conclude that even taking into account a degree of brine mobility, the closed system hypothesis is unable to satisfy all the available chemical and mineralogical data, consistent with the conclusions of Squyres et al. (2006) that scenarios with  $SO_3$ /basalt > 0.25 necessarily imply loss of at least part of the brine component. In



**FIGURE 2.** Kinetic model of alteration with SO<sub>3</sub>/basalt = 0.25 and W/R = 0.6. Panel A represents chemical compositions in the ternary diagram (Ca + Mg + Fe), (Si + Al), S. The red line is the trend of the solid fraction. The blue line is the trend of the coexisting brine. The total simulation time is 20 years and compositions at 5 and 10 years are indicated. The compositions of abraded Meridiani bedrocks are shown as black triangles. The black line connects solids and coexisting brine after ~2 years, a solution which can best explain the trend in bedrock data. Panel B represents the mineralogy of the solid fraction as a function of time. Note that after 2 years significant olivine is present, in contrast to analysis of bedrock.

the case that  $SO_3$ /basalt > 0.25, the pH of the initial brine will be lower (for a given W/R), qualitatively leading to lower stability of olivine and greater stability of sulfates, in better agreement with observations.

Quantitatively, at a W/R of 1, we find that a SO<sub>3</sub>/basalt of 0.48 represents an upper limit to scenarios capable of accounting for Meridiani data because at higher values even the calculated solid fraction is too sulfur rich to be compatible with MER data. For  $SO_3$ /basalt = 0.48 and alteration times >6 years, the chemical composition of the solid fraction shows a trend toward an (Mg,Ca,Fe)SO<sub>4</sub> component (Fig. 3a). In the time range 12 to 20 years, the compositions of the altered solids fall close to Meridiani bulk-rock data with little or no contribution of the brine component necessary (Fig. 3a). Furthermore, during this time interval, olivine has been eliminated, but significant primary pyroxenes and plagioclase remain (Fig. 3b). Secondary mineralogy is dominated by sulfates (jarosite, gypsum, and epsomite), iron oxide, and amorphous silica, all these mineralogical characteristics being consistent with mini-TES and Mössbauer measurements of the rocks at Meridiani Planum (Morris et al. 2006; Glotch et al. 2006). While stressing that a SO<sub>3</sub>/basalt ratio of 0.48 is an upper limit, we conclude that a simple and self-consistent working hypothesis to explain the petrogenesis of rocks analyzed by Opportunity is that they are dominantly the solid residues of in situ alteration of basalt, a significant fraction of the coexisting brines having been transported elsewhere before evaporation. Because of the low water/rock ratio, the removal of the brine exported a small fraction of the rock components without affecting the bulk composition in terms of the ratio of (Si + Al) and (Ca + Fe + Mg) (see Figs. 2a and 3a). Although the final locality of brine evaporation is not immediately apparent, this latter conclusion is broadly consistent with pedogenic models proposed for martian soils (Amundson et al. 2008).

Furthermore, these kinetic calculations predict that continued hydrolysis (resulting from alteration of pyroxenes and plagioclase) acts to neutralize the aqueous solution, destabilizing jarosite and stabilizing phyllosilicates (Fig. 3b). Thus, the close correspondence between modeled and observed mineralogy is



**FIGURE 3.** Kinetic model of alteration with  $SO_3$ /basalt = 0.48 and W/R = 1. Panel A using symbols and colors identical to Figure 2. In this simulation, little or no brine component is needed to reproduce the bulk-rock chemistry of Meridiani samples. Panel B represents the mineralogy of the solid fraction as a function of time. After 20 years the proportion of jarosite decreases, giving way to the precipitation of clays. The stability of epsomite at these conditions has been checked and validated by performing calculations with the Pitzer formalism in the  $SO_4$ -Mg-Ca sub-system.

limited in cumulative time (in this case  $< \sim 20$  years). Restricted cumulative alteration times (presumably controlled by the lifetime of liquid water) are consistent with an emerging picture of conditions at martian surface during the last 3.5 Ga (Chevrier and Mathé 2007; Hurowitz and McLennan 2007; Sefton-Nash and Catling 2008). Even so, the time-scales modeled here are even shorter than those previously proposed based upon hematite concretion growth (390-1900 years; Hausrath et al. 2008) or the dissolution of olivine (370 years; Stopar et al. 2006), but consistent with those reported by Zolotov and Mirenko (2007). On the one hand, it is possible that real time-scales of alteration were somewhat longer than those calculated here, either for the reasons discussed previously, or due to alteration occurring at temperature significantly lower than 0 °C, made possible by important freezing-point depression of concentrated sulfuric acid solutions. On the other hand, extremely short time-scales are to be expected given the very low pH (-0.4) of the initial aqueous solutions when  $SO_3$ /basalt = 0.48 and W/R = 1. However, uncertainty in the time-scale does not invalidate our other principal conclusion that the brine was most probably removed before evaporation.

Finally, the ephemeral nature of liquid water and the highly acidic nature of aqueous solutions predicted here would have made survival of life at that time extremely challenging.

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