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Martian magmatic clay minerals forming vesicles: perfect niches for emerging life?

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Abstract:

Mars has been habitable in its early history but the consensus is that it is quite inhospitable today, in particular because its modern climate cannot support stable liquid water at the surface. Here we report the presence of magmatic Fe/Mg clay minerals within the mesostasis of the Martian meteorite NWA5790, an unaltered 1.3 Ga nakhlite archetypal of the Martian crust. These magmatic clay minerals exhibit a vesicular texture forming a network of micro-cavities/pockets which could serve as micro-reactors allowing molecular crowding, a necessary step for the emergence of life. Because their formation does not depend on climate, such niches for emerging life may have been generated on Mars at many periods of its history, whatever the stability/availability of liquid water at the surface.

1. Introduction

The concept of habitability is essentially tied to the presence of liquid water at (or near) the surface of a planetary body. The surface of Mars has been habitable during the Noachian (4.1 - 3.7 Ga) because liquid water was then both stable and available, at least episodically. Environments likely existed on the Noachian Mars with liquid water and metabolic energy sources available for the development of life (Westall *et al.*, 2013; Grotzinger *et al.*, 2014; Kral *et al.*, 2014; Hurowitz *et al.*, 2017; McMahon *et al.*, 2018). This view relies on the widespread occurrence of Noachian Fe/Mg clay minerals interpreted as products of the alteration of pre-existing silicates by (sub)surface liquid water (Bibring *et al.*, 2006; Ehlmann *et al.*, 2011; Carter *et al.*, 2013; Sun and Milliken, 2015; Viennet *et al.*, 2019). But this may not be the entire story. In fact, some clay minerals can directly precipitate from magmatic fluids (Meunier *et al.*, 2008, 2012; Berger *et al.*, 2014, 2018; Viennet *et al.*, 2020). Because these minerals cannot be used as a proxy for the past presence of liquid water, it seems a priori difficult to consider such magmatic clay minerals as indicators of habitability. Still, as discussed here, magmatic clay minerals may offer fantastic opportunities for prebiotic reactions.

To date, the only occurrence of Martian magmatic Fe/Mg clay minerals has been found in the evolved (alkali/felsic) mesostasis of Nakhla (Viennet *et al.*, 2020), the Martian meteorite eponym for nakhlites. Whether or not these minerals are more than anecdotal on Mars is difficult to determine, especially given the fierce debate over the exact fraction of the Martian crust made up of evolved (alkali/felsic) rocks (Sautter *et al.*, 2016; Udry *et al.*, 2018; Bouley *et al.*, 2020). Here we report the presence of magmatic Fe/Mg clay minerals within the mesostasis of NWA5790, an unaltered Amazonian nakhlite (~1.3-1.4 Ga - Nyquist *et al.*, 2001). In contrast to that of Nakhla,

the mesostasis of NWA5790 does not exhibit a high level of igneous differentiation, making this meteorite more archetypal of the Martian crust (Udry et al., 2018).

2. Materials and Methods

2.1 Section of NWA5790 investigated

The section of NWA 5790 investigated here is the section B described in Jambon et al. (2016). This section was selected because it does not exhibit any trace of terrestrial alteration (Jambon et al., 2016) nor “desert varnish” or “caliche”, i.e. clay mineral sheets or calcite veins produced by alteration (Tomkison et al., 2015; Balta et al., 2017). The augite and olivine grains of the section investigated do not display any iddingsite, amphibole or smectite vein (see figure S1).

2.2 Scanning and Transmission Electron Microscopy

Scanning electron microscopy (SEM) was performed on a thin section of Nakhla using a SEM-FEG Ultra 55 Zeiss (IMPMC - Paris, France) microscope operating at a 15-kV accelerating voltage and a working distance of 7.5 mm for imaging with backscattered electrons and energy dispersive X-ray spectroscopy (EDXS) mapping. Transmission electron microscopy in scanning mode (STEM) was performed on FIB foils using a Thermofisher Titan Themis 300 microscope operated at 300 keV (CCM – Lille, France). TEM-based hyperspectral EDXS data (see below) were obtained using the super-X detector system comprising four windowless silicon drift detectors of high sensitivity. The probe current was set at maximum 200 pA with a dwell time at 10 μ s per pixel.

2.3 Focused ion beam preparations

Focused ion beam (FIB) ultrathin sections were extracted using an FEI Strata DB 235 (IEMN, Lille, France). Milling at low Ga-ion currents minimises common artefacts including local gallium implantation, mixing of components, creation of vacancies or interstitials, creation of amorphous layers, local compositional changes or redeposition of the sputtered material on the sample surface(Wirth, 2009).

2.4 EDXS data processing

A key aspect of this work is the post-processing of the collected EDXS hyperspectral data, performed using the Hyperspy python-based package(De La Pena, Francisco et al., 2017). The signal was first denoised using PCA and then fitted by a series of Gaussian functions. The integrated intensities of the Gaussian functions were used to quantify the spectra thanks to the Cliff-Lorimer method, using experimentally determined k-factors. Special care was taken to correct for absorption effect within the sample, in particular for oxygen X-rays. Absorption correction depends on thickness density, which can be determined by comparing quantification made by Iron L-lines and Iron K-lines(Morris, 1980). Each pixel was quantified independently, and end-member phases were identified based on mixing diagrams.

2.5 Raman spectroscopy

Raman was performed using a homemade time-resolved Raman spectrometer (Beyssac et al., 2017). The laser is a nanosecond pulsed DPSS laser operating at 532 nm with a 1.2 ns duration (FWHM) for the pulse, 10 to 2000 Hz repetition rate and up to 1 mJ output energy per pulse. The laser was focused at the sample surface through a microscope objective (x20, numerical aperture 0.42) and the Raman signal was collected in the backscattering geometry. A Notch filter was

used to cut the Rayleigh scattering. The signal was collected in an optical fiber and sent into a modified Czerny-Turner spectrometer manufactured by Princeton Instrument to be measured using a PIMAX4 ICCD camera manufactured by Princeton Instrument.

3. Results

Like the other nakhlites, NWA5790 is an unaltered cumulate rock exhibiting large euhedral/subhedral crystals of augite that are set in a crystalline mesostasis (Fig 1 and 1S). Consistent with previous reports (Tomkinson *et al.*, 2015; Jambon *et al.*, 2016; Balta *et al.*, 2017), petrographic observations show that the mesostasis of NWA5790 is made of Na/Ca-plagioclases and K-feldspars embedding skeletal Fe-rich Ti-oxides and Ti-rich magnetites hosting ilmenite exsolutions (Fig 1). In addition to augite grains, the investigated section contains an unaltered euhedral olivine (Fig. S1) and a zone of about the same size composed of fibrous minerals (Fig 1). This zone exhibits a radial structure, with a central pore of about 200 μm in diameter, surrounded by Fe/Mg clay minerals intimately mixed with Fe-Si-Al-Ca nanograins, themselves surrounded by dendrites of maghemite and rather amorphous Fe-Si-Al-Ca materials in contact with augite grains and the mesostasis (Fig 1 and supplementary infos). In contrast to the contact with augite grains, the transition with the mesostasis is not sharp, some Na/Ca plagioclases and skeletal equiaxed Fe-rich Ti oxides are distributed within the observed dendrites. These dendrites display a nearly paraboloidal finger-like shape with side-branches, i.e. a columnar dendritic texture with trunks of a few to hundreds of microns in length (Fig 2), typical of sudden cooling/quenching conditions.

The Fe/Mg clay minerals consist of layers of a few hundreds of nanometers in length stacked over a few tens of nanometers, with layer to layer distances of $\sim 11.8 \text{ \AA}$, a typical structure of

smectites (Fig 3). These Fe/Mg clay minerals are chemically heterogeneous, with a mean chemical composition falling between nontronite, celadonite and saponite (Fig 3). Most importantly, the observed anti-correlation between Mg and Cl (Fig. 3) attests to the presence of Cl within the structure of these Fe/Mg clay minerals according to the Mg-Cl crystallographic avoidance principle (Bailey, 1984). This assemblage of Cl-rich Fe/Mg clay minerals is highly porous and exhibits a network of micro-cavities/pockets, from tens of nanometers to several hundreds of nanometers in diameter (Fig 4).

4. Discussion

These Fe/Mg clay minerals have clearly not been produced via the aqueous alteration of pre-existing silicates. In fact, the section of NWA5790 investigated here has been recognised to be unaltered (Tomkinson *et al.*, 2015; Jambon *et al.*, 2016) and the alteration of the Ti-rich and Al-rich mesostasis (mainly made of feldspars) would have produced Ti-rich and Al-rich secondary minerals (Meunier and Velde, 2004). In contrast, the clay minerals described here display a low Al content (4 wt.%) and do not contain any Ti. The Fe and Mg contents of these clay minerals could be consistent with the alteration of augite, but none of the augite grains of NWA5790 displays alteration textures such as retreating surfaces or pitch-like features resulting from dissolution and no contact with the Fe/Mg clay minerals could be observed. Plus, the high Cl content of these Fe/Mg clay minerals is similar to those of Cl-rich magmatic apatites, scapolites and amphiboles found in other nakhlites (Sautter *et al.*, 2006; McCubbin *et al.*, 2013; Giesting and Filiberto, 2016) and to that of the magmatic Fe/Mg clay minerals found in Nakhla (Viennet *et al.*, 2020).

The subhedral/euhedral nature of augite crystals is consistent with slow growth within a magmatic chamber (Treiman, 2005; Jambon *et al.*, 2016; Udry and Day, 2018) or within a chilled margin of a lava flow/sill (Tomkinson *et al.*, 2015). The crystallisation and the accumulation of augite grains led to the entrapment of a residual liquid enriched in Cl, potentially as a result of contamination by an exogenous Cl-rich fluid (McCubbin *et al.*, 2013; Udry and Day, 2018). With decreasing temperature, the Na/Ca plagioclases and the K-feldspars crystallised together with the skeletal equiaxed Fe-Ti oxides and the skeletal Ti-magnetites, eventually leading to the exsolution of a Cl/Fe-rich brine (Fig 1). Here, the screw dislocations at the surface of the augite grains (together with the decrease of solubility of Fe III with decreasing temperature) potentially controlled the very fast precipitation of the observed dendrites, explaining their screw shapes and unidirectional solidification structures (Vernon, 2004). Masses of highly porous Cl-bearing Fe/Mg clay minerals then precipitated from the residual brine (together with or right before the Fe-Si-Al-Ca nanograins), i.e. before reaching a high level of igneous differentiation in contrast to what occurred in Nakhla (Viennet *et al.*, 2020).

The magmatic production of Fe/Mg clay minerals evidenced here in a Martian meteorite archetypal of the Martian crust portends that a possibly significant fraction of the Fe/Mg clay minerals detected on Mars so far may not be the products of the aqueous alteration of pre-existing silicates by (sub)surface water, questioning *a priori* the past habitability of Mars. Yet, the magmatic Fe/Mg clay minerals described here have chemical and physical properties offering fantastic opportunities for (prebiotic) organic reactions (Russell and Martin, 2004; Duval *et al.*, 2020). In fact, in addition to enabling electron transfer, their high Fe content may promote the synthesis and breakdown of universal metabolic precursors (Stucki, 2006), while their high Mg

content may prompt both ribozyme catalysed and non-enzymatic RNA copying reactions (Adamala and Szostak, 2013).

Clay minerals have long been suggested as the perfect means of concentrating organic molecules onto their external surfaces and within their interlayer space so as to be available for prebiotic reactions (Bernal, 1951; Ferris and Ertem, 1992; Brack, 2013; Lagaly *et al.*, 2013; Theng, 2018).

The surface areas of the magmatic Fe/Mg clay minerals described here are very high: external and internal surface areas reach $30 \mu\text{m}^2$ and $670 \mu\text{m}^2$ per μm^3 , respectively (cf supplementary infos). For smectites, this corresponds to 2.4×10^{14} external sites and 5.8×10^{14} internal sites of possible interactions with organic functional groups (cf supplementary infos).

Most importantly, the vesicular texture of these magmatic Fe/Mg clay minerals form a network of chemically heterogeneous micro-cavities/pockets, offering even more optimal conditions for (prebiotic) organic reactions. In fact, most scenarios of the origin of life requires boundaries/membranes (either organic or inorganic) to isolate, concentrate and protect organic compounds that could eventually interact/react with each other (Szostak *et al.*, 2001; Hanczyc, 2003; Chen and Walde, 2010; Sun *et al.*, 2016). Plus, a boundary is a privileged interface for the formation of gradients which can promote organic reactions and could be exploited as an energetic source (Monnard and Walde, 2015; Branscomb *et al.*, 2017; Branscomb and Russell, 2019). Here, more than 600 micro-cavities/pockets can be counted per μm^2 of clay minerals, with surfaces ranging from 1.10^{-5} to $1.4.10^{-2} \mu\text{m}^2$ (Fig 4.C), each of them being potentially a distinct micro-reactor for organic chemistry. The multiple connections between these micro-cavities/pockets may allow organic molecules to diffuse freely into or out these microsystems, possibly leading to molecular crowding, thereby increasing the probability of achieving prebiotic chemical reactions.

5. Conclusion

The section of the Martian nakhlite NWA5790 investigated here contains a zone composed of magmatic Fe/Mg clay minerals mixed with Fe-Si-Al-Ca nanograins surrounded by dendritic maghemite. These magmatic Fe/Mg clay minerals exhibit a vesicular texture forming a network of micro-cavities/pockets which could serve as micro-reactors for the emergence of life. The fact that the formation of such niches for emerging life can be achieved via the magmatic precipitation of Fe/Mg clay minerals should make these aggregates a focus for research into the origin of life, especially as such magmatic Fe/Mg clay minerals form within liquid-water-poor environments (prebiotic reactions are thermodynamically out of equilibrium in aqueous solution - Lambert, 2008). *In fine*, by evidencing the formation of niches for emerging life on Mars even during the Amazonian, the present study provides a strong rationale for the possible emergence of life on other planetary bodies, including rocky and/or icy ones (such as Ceres, Enceladus or Europa) on which the production of clay minerals has recently been reported (Waite *et al.*, 2017; Marchi *et al.*, 2019).

6. References and Notes

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Author Contributions

JCV and SB designed the present study. JCV and SB conducted the SEM experiments. JCV and OB conducted the Raman experiments. JCV, SB and CLG conducted the TEM experiments. JCV and CLG performed SEM and TEM data reduction. All authors contributed to the interpretation of the results. JCV and SB wrote the manuscript, with critical inputs from all authors.

Author Disclosure Statement

No competing financial interests exist.

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Figure Caption

Fig. 1. SEM image of the zone of NWA 5790 investigated in BSE mode (A) and the corresponding EDXS-based mineralogical map (B). Note the euhedral augite grains with no dissolution features surrounding the dendritic maghemite and Fe/Mg clay minerals.

Fig. 2. Dendritic structures. (A) BSE image of the Maghemite showing the location of the FIB foil shown in D-E. (B) Time-resolved Raman spectra of the Maghemite. (C) SEM image highlighting the dendritic texture of the maghemite. (D-E) TEM images in STEM mode of the FIB section showing the screw shape of the dendritic texture along the main direction and their texture in spiral perpendicularly to the main direction.

Fig. 3. Magmatic Fe/Mg clay minerals. (A) SEM image in BSE mode showing the huge mass of clay minerals. (B). SEM image in BSE mode showing the fibrous texture of the clay minerals. (C) TEM image in STEM mode of the FIB section (location shown in Fig. S2) showing the contact between Fe/Mg clay minerals and non-altered silicates. (D) TEM image in bright field showing the classical structure in sheets of the Fe/Mg clay minerals with a d-spacing of 11.7 Å. (E) Diagram showing the chemical composition of the Fe/Mg clay minerals and indicating a mixture of nontronite, celadonite and saponite (left) and plot showing the Mg content as a function of Cl content (right).

Fig. 4. Magmatic Fe/Mg clay minerals. (A) TEM image in STEM mode showing the vesicles formed by the Fe/Mg clay minerals. (B) Size distribution of the vesicles corresponding to the TEM images in STEM mode (E). (C-E). TEM images in STEM mode showing the vesicles formed by the Fe/Mg clay minerals.