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

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16
17 **Abstract:**

18 Mars has been habitable in its early history but the consensus is that it is quite inhospitable today,
19 in particular because its modern climate cannot support stable liquid water at the surface. Here we
20 report the presence of magmatic Fe/Mg clay minerals within the mesostasis of the Martian
21 meteorite NWA5790, an unaltered 1.3 Ga nakhlite archetypal of the Martian crust. These
22 magmatic clay minerals exhibit a vesicular texture forming a network of micro-cavities/pockets
23 which could serve as micro-reactors allowing molecular crowding, a necessary step for the
24 emergence of life. Because their formation does not depend on climate, such niches for emerging
25 life may have been generated on Mars at many periods of its history, whatever the
26 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,

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

54 **2. Materials and Methods**

56 **2.1 Section of NWA5790 investigated**

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

63 **2.2 Scanning and Transmission Electron Microscopy**

64 Scanning electron microscopy (SEM) was performed on a thin section of Nakhla using a SEM-
65 FEG Ultra 55 Zeiss (IMPMC - Paris, France) microscope operating at a 15-kV accelerating
66 voltage and a working distance of 7.5 mm for imaging with backscattered electrons and energy
67 dispersive X-ray spectroscopy (EDXS) mapping. Transmission electron microscopy in scanning
68 mode (STEM) was performed on FIB foils using a Thermofisher Titan Themis 300 microscope
69 operated at 300 keV (CCM – Lille, France). TEM-based hyperspectral EDXS data (see below)
70 were obtained using the super-X detector system comprising four windowless silicon drift
71 detectors of high sensitivity. The probe current was set at maximum 200 pA with a dwell time at
72 10 μ s per pixel.

74 **2.3 Focused ion beam preparations**

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

81 **2.4 EDXS data processing**

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

92 **2.5 Raman spectroscopy**

93 Raman was performed using a homemade time-resolved Raman spectrometer (Beyssac et al.,
94 2017). The laser is a nanosecond pulsed DPSS laser operating at 532 nm with a 1.2 ns duration
95 (FWHM) for the pulse, 10 to 2000 Hz repetition rate and up to 1 mJ output energy per pulse. The
96 laser was focused at the sample surface through a microscope objective (x20, numerical aperture
97 0.42) and the Raman signal was collected in the backscattering geometry. A Notch filter was

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

102 **3. Results**

103 Like the other nakhlites, NWA5790 is an unaltered cumulate rock exhibiting large
104 euhedral/subhedral crystals of augite that are set in a crystalline mesostasis (Fig 1 and 1S).
105 Consistent with previous reports (Tomkinson *et al.*, 2015; Jambon *et al.*, 2016; Balta *et al.*,
106 2017), petrographic observations show that the mesostasis of NWA5790 is made of Na/Ca-
107 plagioclases and K-feldspars embedding skeletal Fe-rich Ti-oxides and Ti-rich magnetites
108 hosting ilmenite exsolutions (Fig 1). In addition to augite grains, the investigated section
109 contains an unaltered euhedral olivine (Fig. S1) and a zone of about the same size composed of
110 fibrous minerals (Fig 1). This zone exhibits a radial structure, with a central pore of about
111 200 μm in diameter, surrounded by Fe/Mg clay minerals intimately mixed with Fe-Si-Al-Ca
112 nanograins, themselves surrounded by dendrites of maghemite and rather amorphous Fe-Si-Al-
113 Ca materials in contact with augite grains and the mesostasis (Fig 1 and supplementary infos). In
114 contrast to the contact with augite grains, the transition with the mesostasis is not sharp, some
115 Na/Ca plagioclases and skeletal equiaxed Fe-rich Ti oxides are distributed within the observed
116 dendrites. These dendrites display a nearly paraboloidal finger-like shape with side-branches, i.e.
117 a columnar dendritic texture with trunks of a few to hundreds of microns in length (Fig 2),
118 typical of sudden cooling/quenching conditions.

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

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

128 129 **4. Discussion**

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

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

157 The magmatic production of Fe/Mg clay minerals evidenced here in a Martian meteorite
158 archetypal of the Martian crust portends that a possibly significant fraction of the Fe/Mg clay
159 minerals detected on Mars so far may not be the products of the aqueous alteration of pre-
160 existing silicates by (sub)surface water, questioning *a priori* the past habitability of Mars. Yet,
161 the magmatic Fe/Mg clay minerals described here have chemical and physical properties offering
162 fantastic opportunities for (prebiotic) organic reactions (Russell and Martin, 2004; Duval *et al.*,
163 2020). In fact, in addition to enabling electron transfer, their high Fe content may promote the
164 synthesis and breakdown of universal metabolic precursors (Stucki, 2006), while their high Mg

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

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

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

174 Most importantly, the vesicular texture of these magmatic Fe/Mg clay minerals form a network
175 of chemically heterogeneous micro-cavities/pockets, offering even more optimal conditions for
176 (prebiotic) organic reactions. In fact, most scenarios of the origin of life requires
177 boundaries/membranes (either organic or inorganic) to isolate, concentrate and protect organic
178 compounds that could eventually interact/react with each other (Szostak *et al.*, 2001; Hanczyc,
179 2003; Chen and Walde, 2010; Sun *et al.*, 2016). Plus, a boundary is a privileged interface for the
180 formation of gradients which can promote organic reactions and could be exploited as an
181 energetic source (Monnard and Walde, 2015; Branscomb *et al.*, 2017; Branscomb and Russell,
182 2019). Here, more than 600 micro-cavities/pockets can be counted per μm^2 of clay minerals,
183 with surfaces ranging from $1 \cdot 10^{-5}$ to $1.4 \cdot 10^{-2} \mu\text{m}^2$ (Fig 4.C), each of them being potentially a
184 distinct micro-reactor for organic chemistry. The multiple connections between these micro-
185 cavities/pockets may allow organic molecules to diffuse freely into or out these microsystems,
186 possibly leading to molecular crowding, thereby increasing the probability of achieving prebiotic
187 chemical reactions.

188

189 **5. Conclusion**

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

203

204 **6. References and Notes**

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341

342 **Author Contributions**

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

347

348 **Author Disclosure Statement**

349 No competing financial interests exist.

350

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356

357 **Figure Caption**

358 **Fig. 1.** SEM image of the zone of NWA 5790 investigated in BSE mode (A) and the
359 corresponding EDXS-based mineralogical map (B). Note the euhedral augite grains with no
360 dissolution features surrounding the dendritic maghemite and Fe/Mg clay minerals.
361

362 **Fig. 2.** Dendritic structures. (A) BSE image of the Maghemite showing the location of the FIB
363 foil shown in D-E. (B) Time-resolved Raman spectra of the Maghemite. (C) SEM image
364 highlighting the dendritic texture of the maghemite. (D-E) TEM images in STEM mode of the
365 FIB section showing the screw shape of the dendritic texture along the main direction and their
366 texture in spiral perpendicularly to the main direction.
367

368 **Fig. 3.** Magmatic Fe/Mg clay minerals. (A) SEM image in BSE mode showing the huge mass of
369 clay minerals. (B). SEM image in BSE mode showing the fibrous texture of the clay minerals.
370 (C) TEM image in STEM mode of the FIB section (location shown in Fig. S2) showing the
371 contact between Fe/Mg clay minerals and non-altered silicates. (D) TEM image in bright field
372 showing the classical structure in sheets of the Fe/Mg clay minerals with a d-spacing of 11.7 Å.
373 (E) Diagram showing the chemical composition of the Fe/Mg clay minerals and indicating a
374 mixture of nontronite, celadonite and saponite (left) and plot showing the Mg content as a
375 function of Cl content (right).
376

377 **Fig. 4.** Magmatic Fe/Mg clay minerals. (A) TEM image in STEM mode showing the vesicles
378 formed by the Fe/Mg clay minerals. (B) Size distribution of the vesicles corresponding to the
379 TEM images in STEM mode (E). (C-E). TEM images in STEM mode showing the vesicles
380 formed by the Fe/Mg clay minerals.
381







