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Advanced two- and three-dimensional insights into Earth's oldest stromatolites (ca. 3.5 Ga): Prospects for the search for life on Mars

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ABSTRACT

Paleoarchean stromatolites are among the oldest compelling evidence for life. We present advanced two- and three-dimensional (2-D and 3-D) reconstructions of the morphology, mineralogy, trace element geochemistry, and taphonomy of permineralized stromatolites from the lowermost horizons of the ca. 3.5 Ga Dresser Formation, Pilbara, Western Australia. Rare earth element plus yttrium compositions suggest a restricted paleodepositional setting influenced by marine influxes; this contrasts with other Dresser stromatolites, which developed around terrestrial hot springs. Mineral phase relationships and positive Eu anomalies denote syndepositional hydrothermal influence and silicification promoting high-fidelity microstructural preservation. Although no primary kerogen is preserved, numerous 2-D and 3-D morphological characteristics denote a biogenic origin, including the onlap of sedimentary layers onto stromatolitic topography, fine-scale undulatory laminations, non-isopachous laminations with crestal thickening, laminoid fenestrae, and subvertical pillar-like fabrics interpreted as microbial palisade structure; these features suggest that the stromatolite ecosystem was dominantly phototrophic. The deep iron-rich weathering profile of the Dresser stromatolites makes them pertinent analogues for potential microbialites in altered carbonates on Mars. Were similar putative biogenic macro-, meso- and micromorphologies identified in habitable Martian settings by rover imaging systems, such materials would be compelling targets for sample return.

INTRODUCTION

Microbialites reflect interactions between the ecology and ecophysiology of benthic microbial communities, sedimentation, mineral precipitation, hydrodynamics, and ambient geochemistry (Bosak et al., 2009). Their fossils provide an almost 3.5-billion-year archive of biospheregeosphere co-evolution; indeed, stromatolites from the 3.43 Ga Strelley Pool Formation, Western Australia, represent the oldest widely accepted traces of life (Allwood et al., 2009,

2010). Slightly older probable stromatolites have been described from the 3.48 Ga Dresser Formation at North Pole Dome, Western Australia (Dunlop et al., 1978; Walter et al., 1980; Hickman-Lewis et al., 2017, 2019; Fig. 1; Figs. S1 and S2 in the Supplemental Material¹). Within the same horizons of the Dresser Formation, microfossil-like objects, negative $\delta^{13}C_{org}$, carbonaceous materials rich in aliphatic moieties and other functional groups, and $\delta^{34}S$ systematics further indicate diverse microbial ecosystems (Shen et al., 2001; Ueno et al., 2006; Philippot et al., 2007; Noffke et al., 2013; Igisu

et al., 2018; Mißbach et al., 2021). Nonetheless, a great burden of proof falls upon establishing the biogenicity of such ancient traces of life, and the veracity of Dresser stromatolites and microfossils has been challenged (Lowe, 1994; Wacey et al., 2018; Bosak et al., 2021).

Stromatolitic strata in the Dresser Formation are associated with vein swarms (Fig. 1A) comprising black chert + organics \pm barite \pm pyrite \pm quartz (Tadbiri and Van Kranendonk, 2020) and are interlayered with bedded barite (Figs. 1B and 1C). Local cross-cutting veins suggest syndepositional hydrothermal activity (Nijman et al., 1999; Van Kranendonk and Pirajno, 2004; Harris et al., 2009). Bedded barite (Fig. 1B) has been suggested to arise from either hydrothermal activity (Nijman et al., 1999) or the replacement of gypsum precursors (Buick and Dunlop, 1990). Previous descriptions of Dresser stromatolites identified wrinkly, coniform, domical, and pseudocolumnar macrostructures (Walter et al., 1980; Buick et al., 1981; Van Kranendonk et al., 2018). Because metamorphism at North Pole Dome does not exceed greenschist grade (Hickman, 2012) and because similar biogenic stromatolite forms occur throughout the geological record, these structures cannot arise exclusively from deformation. The Dresser Formation is, thus, crucial for understanding the earliest ecosystems on Earth and is potentially a key analogue in the search for life on Mars.

In this study, we conduct a multi-technique characterization of microstructures in Dresser

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¹Supplemental Material. Methods, Figures S1–S16, and Table S1. Please visit https://doi.org/10.1130/GEOL.S.21200434 to access the supplemental material, and contact editing@geosociety.org with any questions.

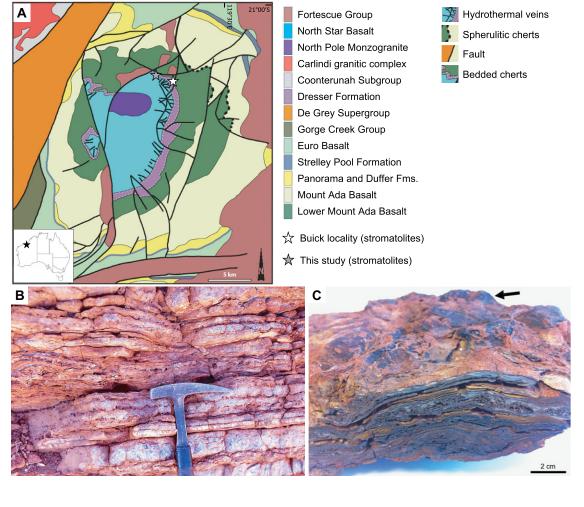


Figure 1. Context and morphology of Dresser Formation (Pilbara, Western Australia) stromatolites. (A) Geological map of the North Pole Dome locality. (B) Outcrop photograph showing finely layered stromatolites interbedded with chert-barite. (C) Hand sample with domical topography (arrowed).

Formation stromatolites, establishing their biogenicity and exploring their potential as taphonomic analogs for putative Martian biosignatures.

MATERIALS AND METHODS

Dresser stromatolite samples were obtained from 1- to 1.5-m-thick beds within the lower-most horizons of the North Pole Dome chert-barite unit overlying a volcanic conglomerate (lithostratigraphic assemblages A1 and A2 of Djokic et al., 2021; Fig. 1; Fig. S2). These stromatolites comprise millimeter- to centimeter-scale stratiform and domical layers intercalated with silica and barite (Figs. 1B and 1C).

Samples were prepared as polished small-format $(27 \times 46 \text{ mm})$ and large-format $(51 \times 75 \text{ mm})$ thin sections and cylindrical samples of 2–8 mm diameter. We performed millimeter- to submicron-scale correlative micro-analysis using optical microscopy, Raman spectroscopy, laser ablation inductively coupled plasma—mass spectrometry (ICP-MS), and laboratory and synchrotron computed tomography (see the Supplemental Material for methods) in order to elucidate 2-D and 3-D morphological complexity, phase relationships, major and trace element geochemistry, and taphonomy at the highest possible resolution.

RESULTS

Mineralogy and Lithochemistry

Laser ablation ICP-MS 60 μ m point analyses in syndepositional microcrystalline chert show enrichments in heavy rare earth elements (REEs) relative to light REEs, mostly positive La/La* (0.21–22.0; median = 1.55), Y/Y* (0.58–62.6; median = 1.35), and Gd/Gd* (0.84–4.14; median = 1.48) anomalies and strongly positive Eu/Eu* anomalies (3.20–58.4; median = 12.8) (Fig. 2A; see the Supplemental Material). Y/Ho ratios range from 18.2 to 213.1 (median = 40.9).

Raman spectroscopy and scanning electron microscopy with energy dispersive X-ray analysis (SEM-EDX) identified three mineral phases: hematite, barite, and quartz (Figs. 2B–2F; Figs. S6–S11). High-resolution SEM-EDX maps show micron-scale intermixing of the constituent mineral phases, particularly barite and quartz (Figs. 2D–2F).

2-D and 3-D Morphologies

Dresser stromatolites are characterized by stratiform and domical macrostructures (Figs. 1B and 1C). Optical microscopy and laboratory X-ray micro-computed tomography (μ CT) (voxel resolution = 6.5 μ m) reveal mil-

limeter- to centimeter-scale laminated domes dominantly composed of hematite and silica, and thicker layers comprising mostly barite and silica (Figs. 3 and 4; Fig. S12). Synchrotron μCT (voxel resolution = $0.9 \mu m$) also shows that the outer veneer of the samples comprises dense iron oxide (Fig. 4D; Fig. S16). Higher-magnification ($200 \times -1000 \times$) optical microscopy shows a microstructure comprising undulatory \sim 30 μ m laminations (Fig. 3B). Hematitic laminations are laterally non-isopachous, commonly thickening within the crests of domical structures (Figs. 3C, 3D, 4A, and 4C); barite-silica laminations are, by contrast, mostly isopachous (Fig. 3A) and onlap onto domical hematitic structures in some cases (Fig. 3E). Several layers feature distinct subparallel alignments of hematitic fabric elements oriented perpendicular to macrostructural laminations and separated by void spaces (Fig. 3C). These aligned pillarlike fabrics are identifiable in synchrotron µCT reconstructions, forming a 3-D field of microcolumnar and/or pillar-like features that extend laterally over several millimeters (Fig. 4E). Ragged and irregular void spaces mostly occur in hematite layers (Figs. 2B, 2D, 2F, and 3A) and are also visible in 3-D visualizations (Figs. 4B, 4D, and 4E).

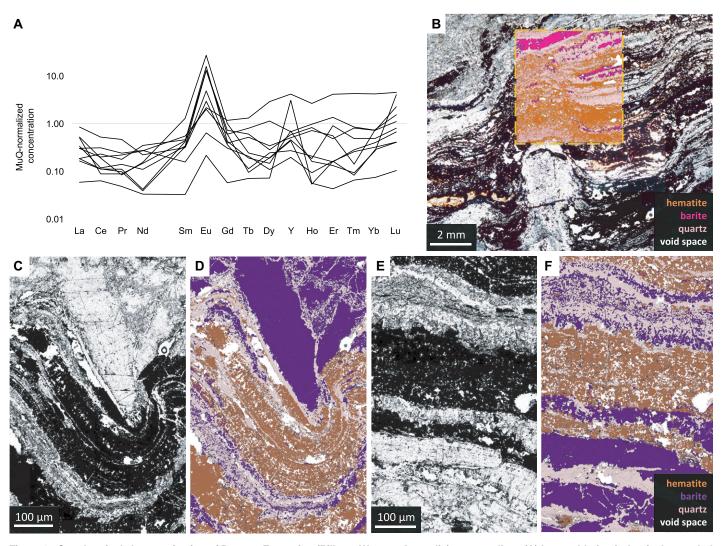


Figure 2. Geochemical characterization of Dresser Formation (Pilbara, Western Australia) stromatolites. (A) Laser ablation inductively coupled plasma—mass spectrometry (LA-ICP-MS) analyses from chert normalized using Mud of Queensland (MuQ). (B) Raman mineralogy mapping of domical stromatolite. (C–F) Scanning electron microscopy with energy dispersive X-ray analysis (SEM-EDX) phase maps of stromatolites.

DISCUSSION

Our data provide the first comprehensive 2-D and 3-D morphological and geochemical appraisal of Earth's oldest stromatolites, which, despite severe recrystallization and weathering, archive records of paleoenvironment, paleoecology, and taphonomy. Coupling thin section and microtomographic data sets facilitates heretofore unprecedented 2-D and 3-D reconstructions of stromatolitic microstructure.

Paleoenvironment

Formerly interpreted as having been deposited in an evaporative marine setting (Dunlop et al., 1978; Walter et al., 1980), subsequent studies re-evaluated the Dresser Formation as a shallow-water, low-eruptive caldera lagoon deposit associated with hydrothermal circulation driven by the underlying magma chamber (Nijman et al., 1999; Van Kranendonk and Pirajno, 2004). Nonetheless, marine influence has been noted in North Pole Dome chert-barite

sequences (Dunlop et al., 1978; Van Kranendonk et al., 2003), indicating that seawater routinely intermixed with hydrothermal fluids flowing through the lagoon via connections with the open ocean. Syndepositional quartz analyzed herein exhibits slight heavy REE enrichment and positive La, Y, and Gd anomalies consistent with marine influence, while strongly positive Eu anomalies signify hydrothermal influence (Allwood et al., 2010). While several recent studies have suggested a subaerial hot spring setting for certain Dresser stromatolites (Van Kranendonk et al., 2018; Djokic et al., 2021), our in situ geochemical analyses more closely align with the long-established interpretation of a hydrothermally influenced shallow-marine lagoonal environment (Dunlop et al., 1978; Van Kranendonk et al., 2003; Noffke et al., 2013). In particular, our chert REE + Y patterns are distinct from those in Djokic et al. (2021), which are more consistent with a terrestrial setting; however, Djokic et al. performed whole-rock

analysis, not in situ study as conducted herein. In situ analyses of heterogeneous Precambrian microbialites have the advantage of linking geochemical signatures to specific carrier phases while facilitating detection of anomalous and/or contaminative signals.

Tapering barite morphologies reminiscent of bottom growth crystals suggest that some detrital barite arises from precursor gypsum (cf. Buick and Dunlop, 1990); however, field and textural relationships with silica also suggest that some barite is syndepositional, of hydrothermal genesis (Van Kranendonk et al., 2018). We therefore advocate for combined marine and hydrothermal influence over primary mineralogy.

Biogenic Fabrics

Although no kerogen is preserved, 3-D morphological complexity and variation in the studied samples is strikingly consistent with biogenic origins. Sedimentary chert-barite

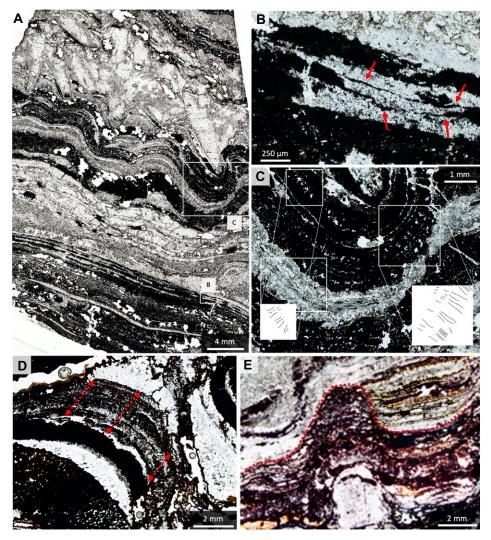


Figure 3. Optical photomicrographs showing ensemble of biogenic fabrics. (A) Thin section showing alternating undulating and planar hematite layers (dark layers) interspersed with chert-barite (light regions). (B) Fine-scale undulatory laminations. (C) Pillar-like "palisade" fabric; sketch plan shows distribution of fabric elements. (D) Crestal thickening in laminations (arrows). (E) Onlap of sedimentary laminations (yellow) against stromatolite dome (red).

layers onlapping onto domical macrostructures suggests microbialites attempting to outpace sediment accumulation (Allwood et al., 2009). The finest laminations (<30 μ m) are of approximately equal dimensions to those of Archean and younger microbialites (Noffke, 2010; Hickman-Lewis et al., 2018), and the thickening of laminations toward crests in the macrostructure may denote trophotactic behavior (Jogi and Runnegar, 2005). Non-isopachous laminations and a lack of inheritance between successive laminations suggest 3-D variations in biological factors including productivity, nutrient diffusion, and gas production, or nondeterministic interactions between microbes and the environment (Hickman-Lewis et al., 2019); the 3-D morphology of laminations is, thus, inconsistent with static inorganic precipitation. We interpret subparallel aligned pillar-like hematitic elements (Fig. 3C) as biogenic palisade structure; of note is that these elements intercalate with flat-laminated horizons to form a layered structure similar in morphology and dimensions to modern microbial palisade (Gong et al., 2020; Álvaro et al., 2021). Synchrotron tomography reveals 3-D networks of hematite-replaced palisade-like structures interspersed with void spaces; this laterally extensive 3-D texture also mirrors that of younger fossilized stromatolites but differs from that of abiogenic precipitates (Allwood et al., 2010). Biogenic palisade structure denotes trophotaxis, which was likely dominantly phototrophic given the shallow paleodepositional depths. Comparable fabrics in sinter deposits from other Dresser horizons have been interpreted similarly (Djokic et al., 2021) but remain to be characterized at high resolution.

Irregular and ragged voids likely represent laminoid fenestrae; in other Paleoarchean stromatolites, such structures are interpreted to form by degassing and/or desiccation of decaying organic materials (Allwood et al., 2010). In contrast, biogenic gas production fenestrae would be rounded, blister or hourglass shaped and account for a large proportion of their host layer (Bosak et al., 2009), deriving from "expansive" growth rather than decay.

Taken together, this ensemble of morphological data affirms the biogenicity of Earth's oldest known stromatolite assemblage. Our approach demonstrates the power of submicron correlative 2-D and 3-D study in establishing stromatolite formation, implying that such techniques should be applied to less intensively studied Archean examples.

Microbialite Taphonomy and Relevance to the Search for Life on Mars

Based on observations of core samples and outcrops, fossilized Dresser stromatolites originally comprised silica, pyrite, carbonate, and barite (Buick and Dunlop, 1990; Van Kranendonk et al., 2018). During metamorphism, silica underwent Ostwald ripening to form microquartz; its REE + Y compositions (Fig. 2A), consistent with other Archean examples, imply that it reflects a primary phase. As outlined above, we find evidence for both primary barite and barite as a replacement phase of precursor gypsum. Stromatolites were likely originally pyritized as part of a well-developed sulfur cycle (Shen et al., 2001; Philippot et al., 2007) and oxidized to hematite (Gu et al., 2020) under recent aridtropical weathering; indeed, Dresser outcrops are oxidatively weathered to a depth of tens of meters (Philippot et al., 2009). This later hematite, though clearly biofabric retentive, does not yield any geochemical information relevant to stromatolite biogenesis.

In terms of their age, likely microbial consortia, and taphonomy, the Dresser stromatolites represent compelling biosignatures for Mars exploration. Due to long-term oxidative conditions at the surface of Mars, similar oxidative weathering phases may be expected to coat and pervade putative microbialites in once-habitable Noachian terranes; indeed, most abraded rocks on Mars show hematite-rich coatings overlying relatively well-preserved protoliths (Edgett et al., 2015). Such materials might also be expected to be kerogen-poor in their uppermost centimeters to meters but nonetheless be valuable morphological biosignature repositories.

Microbialites are prime biosignature targets for the Mars 2020 Perseverance rover mission. Their macrostructure (e.g., Fig. 1) could be visible in Remote Micro-Imager and Mastcam-Z images at outcrop scale whereas fine-scale features (tens to hundreds of microns in size) should be identifiable in images from the WAT-SON camera. Nested WATSON imaging at high resolution (from $\sim\!100$ to $\sim\!10$ $\mu m/pixel)$

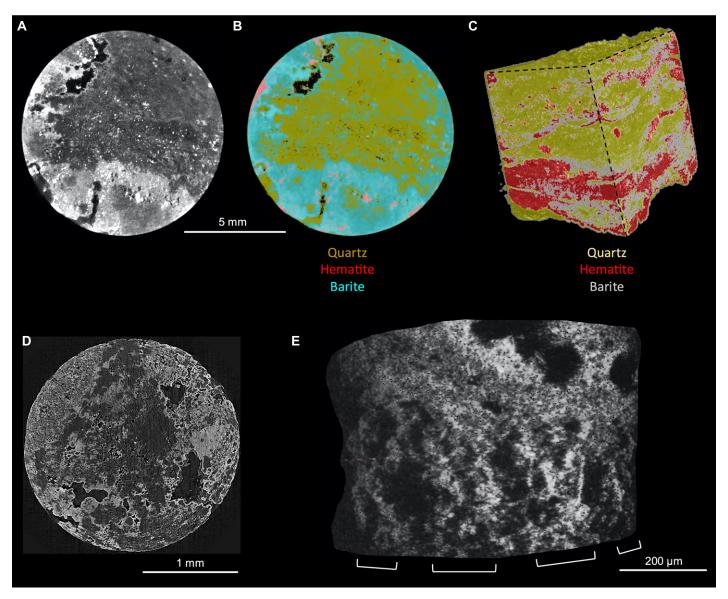


Figure 4. Three-dimensional visualizations of Dresser Formation (Pilbara, Western Australia) stromatolites. (A,B) Laboratory X-ray micro-computed tomography (μ CT) tomogram. (C) Laboratory X-ray μ CT (voxels = 6.5 μ m) reconstruction showing domical stromatolite morphology. (D) Synchrotron μ CT tomogram indicating density distributions within stromatolitic microstructure. (E) Synchrotron μ CT rendering (voxels = 0.9 μ m) of hematite (gray) preserving palisade structure formed of pillar-like features (bracketed) separated by voids (dark regions).

could identify macrostructure, mesostructure, and much microstructure. If coupled with geochemical data using the payload instrumentation (Farley et al., 2020), a paleodepositional and taphonomic history could be deduced. Nonetheless, describing stromatolitic microstructure and unambiguously overcoming the burden of proof on the biogenicity of such ancient materials would likely require sample return and advanced multi-technique analysis such as described herein. The case of the Dresser stromatolites allows us to propose that although putative biofabrics in heavily altered samples should be considered with caution and rely upon correlative 2-D and 3-D techniques, the fact that numerous biogenic characteristics can be imaged in such ancient and altered samples bodes well for the identification of morphological biosignatures in Martian materials, both in *situ* on Mars and following sample return.

CONCLUSIONS

The studied Dresser Formation stromatolitic horizons were deposited in a shallow lagoon setting influenced by marine and hydrothermal fluids. Microbial consortia responsible for constructing these stromatolites would have been phototrophic, thermophilic, and likely metallophilic due to the liberation of metal ions from local basalts. Early and rapid silicification preserved fine-scale 3-D biogenic micromorphologies despite subsequent alteration. Our high-resolution correlative micro-analyses cement the Dresser Formation as a key horizon in the search for Paleoarchean ecosystems.

Occurring within the stratigraphically lowermost sedimentary horizons of the Dresser Formation, these stromatolites are the oldest direct evidence for life on Earth. Their paleodepositional setting, polyextremophilic biology, and taphonomy make them ideal analog biosignatures for Mars, reflecting the type of morphological fossils one might expect to encounter in altered Noachian carbonates.

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Australia in 2000, prior to the establishment of this locality as the Buick Geoheritage Reserve in 2010. We thank Sylvain Janiec (Institut des Sciences de la Terre d'Orléans) for sample preparation, and Vincent Fernandez and Callum Hatch (UK Natural History Museum) for technical support. 3-D visualizations were generated using Dragonfly (https://theobjects.com/dragonfly). Two reviewers provided constructive comments

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