Solving the mystery of desert varnish with microscopy

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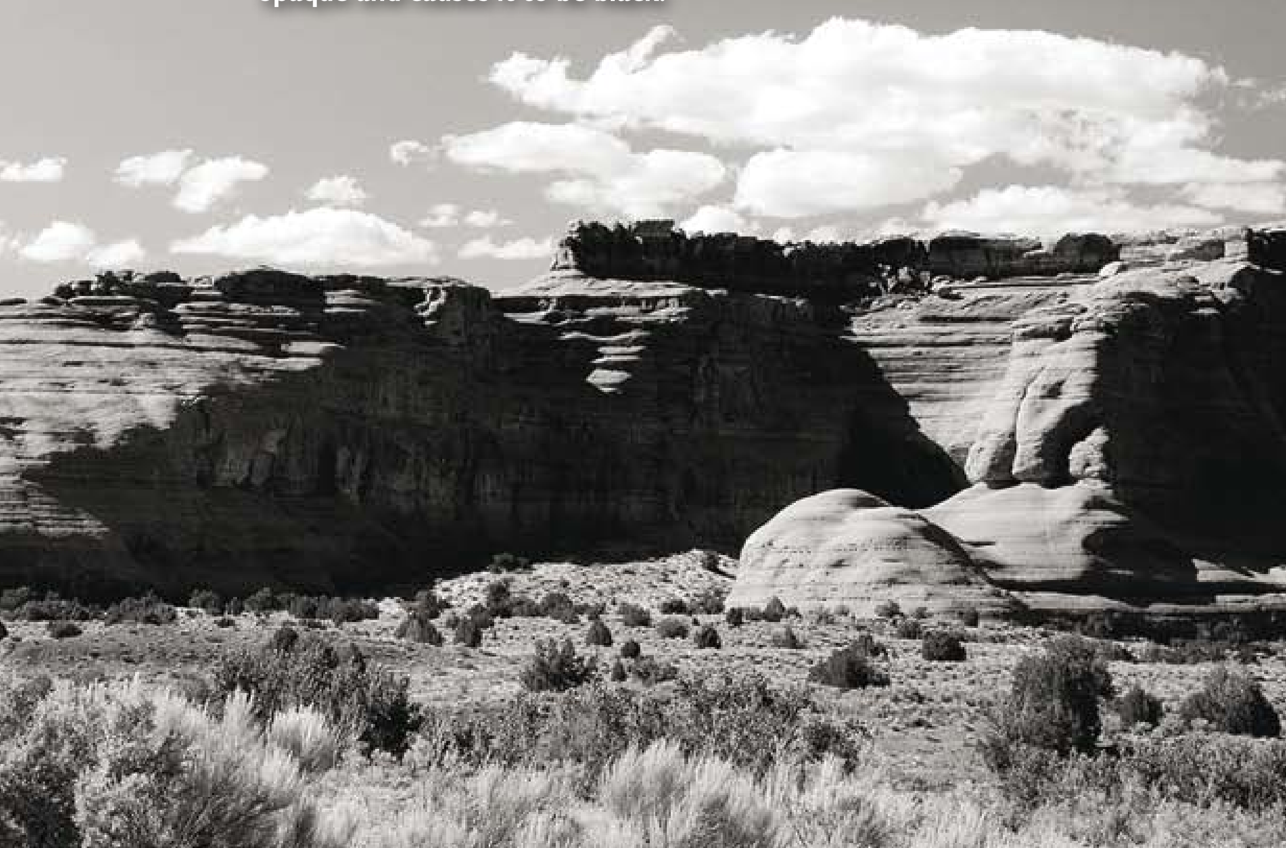
In areas such as Death Valley California whole mountains shimmer as light is reflected from widespread coatings of black opalescent desert varnish (Figure 1). Similar desert varnishes have been found on all continents, in locations such as the Gobi (Figure 2), Sonoran, Mojave, Namibian (Figure 3), Victorian and Atacama Deserts. These dark, lustrous coatings have attracted the interest of scientists for centuries. In 1852, the German naturalist and explorer Alexander Humboldt observed desert varnish on a transatlantic expedition and questioned how this enigmatic feature may have formed. His contemporary, Charles Darwin also engaged in the search for explanations for this unusual rock coating and, in 1871, attempted to satisfy his interest by performing chemical analyses. To date many other noteworthy scientists have examined desert varnish and have commented on its bulk chemistry, the arid conditions in which it forms in and the concentration of manganese that makes it opaque and causes it to be black.



Humboldt's questions are nearly identical to the ones that we ask 150 years later

Fig. 1. Death Valley California desert varnish.

Fig. 2. Desert varnish coating rocks in a stone pavement, Gobi Desert, Mongolia.

Fig. 3. Desert varnish on a rock slope in Namibia. Picture contributed by W. Krumbein and A. Gorbushina.

Despite centuries of scientific effort, the origin of desert varnish is still shrouded in controversy. Most investigators have looked to biological causes where microbes create varnish and preferentially concentrate manganese relative to the local soils and rocks. More modern theories propose a non biological origin with sequential episodes of inorganic chemical reactions dominating formation processes.

Finally, microscopy appears to be solving the riddle of desert varnish. Optical studies have provided the first clues that it is not just an unstructured opaque oxide coating but rather a micron scale banded material, reflecting layer upon layer of chemical deposition. Electron microscopy procedures with X-ray analyses have revealed the silica rich nature of these layers and may be finally answering the questions posed by Humboldt, Darwin and subsequent generations of scientists puzzled by this unusual feature of arid environments.

Historical desert varnish studies

The rock coatings that Darwin (1887) described and Humbodlt observed are common throughout the world's arid lands but, until recently, the question of their origins remained unresolved. Sophisticated analytical instruments such as scanning transmission electron microscopes (STEM) and focused ion beam (FIB) have only become available recently. Early efforts did, however, provide clues to desert varnish origin. For instance, Humboldt began to recognize the role of silica when he described granite boulders near Santa Barbara and stated, "The black crust is 0.3 of a line of thickness, it is found chiefly on quartzose parts. The crystals of feldspar sometimes preserve externally their reddish-white colour, and rise above the black crust." Moreover he became the first to conclude that the black crusts are coatings and not weathering products when he showed that breaking the stone with a hammer revealed no evidence of decomposition of the underlying white rock.

Humboldt's summary of his thoughts and his questions are nearly identical to the ones that we ask 150 years later, including, do coatings include concentrated manganese and ferriginous metals and are components added from the atmosphere?

Layers in desert varnish

Desert varnish is a thin sedimentary deposit (~<200 μm thick) and using the microscope its most notable feature is the presence of micron-sized layers. In typical geological thin sections (30 μm thickness) cut normal to the varnish surface the coatings are opaque. The making of a special ultra-thin section revealed structure within the coating. This heterogeneity was first observed by Perry and Adams (1978) who showed that desert varnish coatings are composed of alternating light and dark layers (Figure 4). Silicon and oxygen are the primary elements in desert varnish, but oxides including aluminum, manganese, iron, titanium and magnesium are also important and it is the variability in abundance of these oxides that creates the layers. Dark layers within varnishes are enhanced in manganese by orders of magnitude over the surrounding rocks and soils. Light layers have lower concentrations of black oxides. Less laterally uniform variations in composition are also evident and whole detrital grains can be embedded in varnish coatings. The thickness of layers is also variable and in three dimensions the growth of layers can lead to botryoidal structures (external forms resembling a bunch of grapes) (Figure 4).

Fig. 4. Optical microscope image of ultra-thin section (~10 μm) of varnish coating taken in 1978 (Perry, 1979). White arrows point to abundant trapped detrital grains. The botryoidal structures are in several orientations. Dark areas are regions of enriched oxides, particularly manganese in this sample. Scale bar 20 μm.

Types of rock coating

The black desert varnish is not the only type of rock coating draped in controversy; other coating types have also been observed in nature. A unique characteristic of desert varnish stones is a bright red coating on their undersides (Figure 5). The red bottom-coatings exhibit the same lustrous and shiny appearance of the black top-coatings. They are most often found on smaller stones in playas such as in the Gobi Desert (Figure 2) and in the Mojave Desert (Figure 5).

Silica glazes are also found throughout the world and have been investigated in Hawaii (Curtis *et al.*, 1985 and Farr and Adams, 1984), Morroco (Smith and Whalley, 1988), Oregon (Farr, 1981) and Antarctica. The shiny glazes on rocks including Antarctica were thought to be a result of wind polishing, but recent closer examination of these however shows that they too are coatings (Figure 6). Their clear quality is a result of reduced tinting by oxides. A few weight percent oxides will colour coatings and make them black or red depending on the presence of black manganese or red iron oxides but even these small amounts are not present in most glazes.

Sources of materials

Lustrous rock coatings are observed globally in desert environments and it is widely agreed that desert varnish and silica glazes are true coatings rather than a weathering product of an underlying material (Figure 6). Consequently,

Fig. 5. The glossy red under-glaze, typical of smaller stones, covers the under sides of both opaque and translucent stones.

some source materials for desert varnish must originate from external environments and are introduced by atmospheric transport. Specifically, dust can land on rock surfaces and its constituents take part in chemical reactions with some components becoming concentrated and adhered or "glued" together. This is especially true when lots of water is present (Figure 7). Atmospheric deposits of trace-elements also are introduced and become part of varnish coatings (Thiagarajan and Lee, 2004). Importantly, the chemical composition of desert varnish necessitates that unused elements and materials need to be removed in a never-ending interchange that allows for the concentration and formation of mineral components.

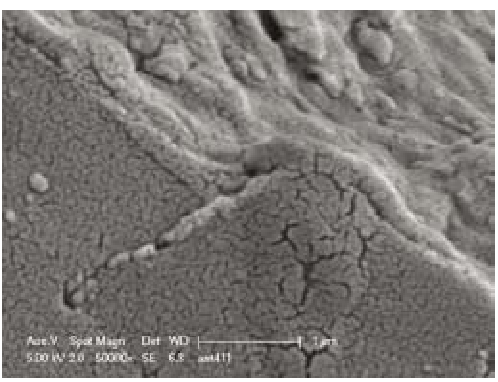


Fig. 6. A polished thin section of very thin Antarctica silica glaze using high magnification SEM (50,000x).

The role of biology

Conventionally the enigmatic organic-rich desert varnish has been ascribed to the action of biology on rock surfaces. Microbes are thought to be pervasive in all environments on Earth and researchers have actively sought a causative link to desert varnish. However, ongoing Martian analogue studies in extreme arid conditions such as the Atacama Desert show that bacteria are rarely present on rock surfaces. In previous times it was suggested that desert varnish was formed by colonies of bacteria living on the rock surface. The black top coatings were attributed to biological action and early investigators speculated that the manganese oxidizing bacteria concentrated the manganese oxides on the top but did not grow as readily underneath rocks. Primarily, these investigations used culture-based studies of microbes, both bacterial and fungal (Hungate *et al*., 1987). The emphasis was on an exhaustive search

Fig. 7. Mojave Desert rock after a rain showing water held in depressions. An ephemeral chemical cauldron in which silicic acid chemistry creates the starting materials for desert varnish.

for manganese and iron oxidizing microbes. The manganese and iron were thought to have been absorbed from dust by the bacteria and oxidized during biochemical reactions over thousands of years (Dorn and Oberlander, 1981). According to

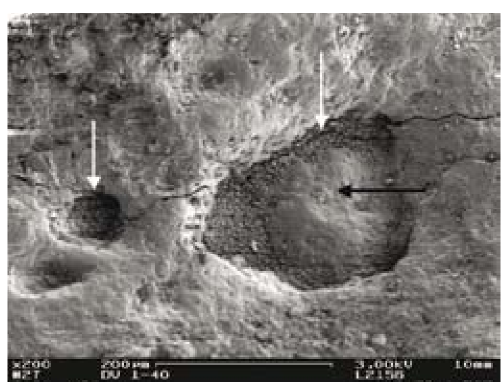


Fig. 8. Botryoidal varnish formation in a depression on the rock surface (white arrows). Note that there is no characteristic mounded texture in areas that are higher than the "dimple" (depicted by the black arrow). The smooth areas were later found to be silica coatings. This sample is from the Panamint Springs, Death Valley, California.

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conventional thought, the silicate particles were also obtained from the atmosphere and cemented together by oxides produced by bacteria. Yet, biological mechanisms remain unsatisfactory and no viable hypothesis has been presented that provides an explanation for key varnish features, such as its hardness, a mechanism for binding the heterogeneous components together, a means of producing the lamellar and botryoidal morphologies (Figures 8 and 9) and its slow rates of formation (Liu and Broeker, 2000). Figure 10 shows a laboratory produced silica desert varnish-like coating made without bacteria (Perry *et al.*, 2005).

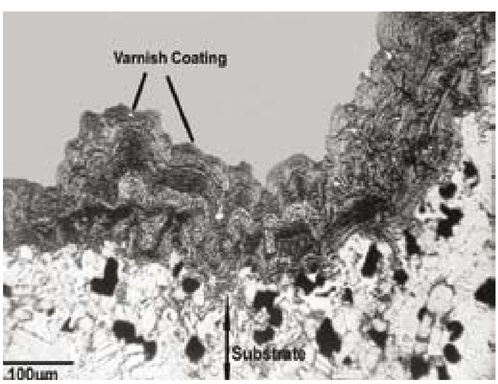

Fig. 9. Ultra – thin section ~10μm think of Sonoran Desert Varnish showing botryoidal mounds and the rock it is covering (Perry and Adams, 1978).

Many forms of biology utilize and concentrate silica including plants and, for example, diatoms c.f. (Perry *et al.*, 2007). Microbes appear to use silica as a UV shield in high elevation hot-springs in Chile and perhaps the mechanism was used by early cyanobacteria (Phoenix *et al.*, 2006).

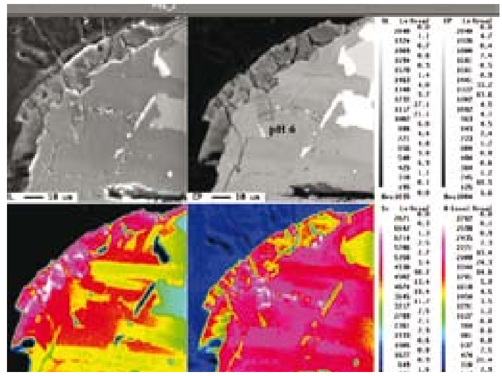

Fig. 10. Silica-rich desert varnish-like coating. The substrate material is fine grained basalt. The electron microprobe X-ray maps were made of coatings formed in the lab at various pH (Perry et al., 2005). The simple addition of manganese can make these layered artifical coatings black.

Life's entombed remains

While cultures of bacteria were obtainable from the surfaces of varnish coated rocks, a direct look at those surfaces using electron microscopes, after fixing the surfaces in order to preserve any biology present, rarely revealed the presence of bacteria. This was revealed by Jones (1991) in the Atacama and Smith and Whalley (1988) in the Atlas Mountains of Morocco and Perry *et al.* (2007) in the Mojave Desert. Yet, it must be noted that during one such study a unique surface fungus was identified by Perry (1979) on the surfaces of varnish coated rocks in the Sonoran Desert

Fig. 11. SEM of desert varnish surface with MCF (micro colonial fungi).

(Figures 11 - 12) and it was later classified by Staley *et al.* (1982) and called micro colonial fungi (MCF). It is an ascomycetes and is visible to the naked eye as a tiny black object. MCF has melanin pigments making it highly UV resistant and it seems to

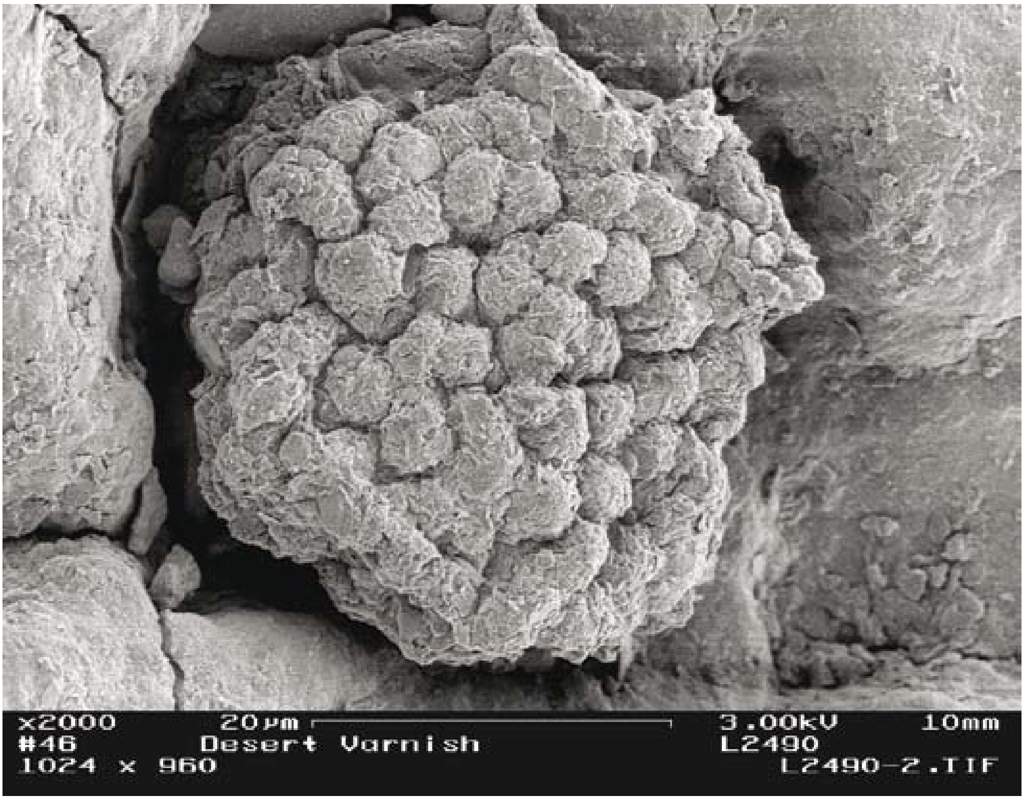

Fig. 12. MCF growing in dimple on the surface of a varnish coated rock.

favour growth in similar niche areas where desert varnish forms (Figure 13). The association was investigated by several scientists and no direct link was found between MCF and desert varnish formation: rather SEM microscopy shows that when the MCF die their residues are captured in desert varnish coatings. Unsuccessful searches for bacteria, therefore, revealed the fundamental mechanism for incorporating the remains of all microbes, some of which were highly exotic. Other organisms deposited on desert varnish surfaces may also become part of coatings but have not been revealed conclusively in micrographs. Figures 14 and 15 show SEM images of a probable pollen and a diatom.

Molecules and varnish

Often, less than complete remains of organisms are

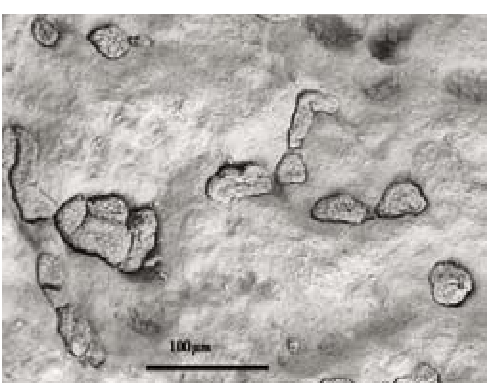

Fig. 13. SEM backscatter image of MCF on varnished rock from the Gobi Desert, Mongolia. Heavier elements are lighter gray. Dark areas around MCF are where X-rays are blocked. MCF pictured vary in size from~30-80µm in diameter.

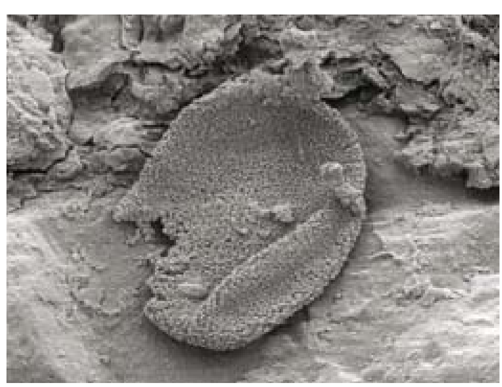

Fig. 14. Probable pollen found on desert varnish ~10µm. SEM image x5000.

Often, less than complete remains of organisms are entombed in desert varnish.

entombed in desert varnish. The first examination of organic molecules being sequestered or complexed in varnish coatings was published by Perry *et al.* (2003). Amino acids were found in coatings and data are shown in Figure 16. The finding of unexpectedly large amounts of labile serine was at first perplexing. However, after considering that amorphous silica (figure 17) is important in desert varnish formation it was found that the hydroxylated serine's OH (Figure 18) is nearly the same distance as the two OH hydroxyls in silicic acid (approximately 26 nm). Other organic compounds may make a C-O bond or be entombed in the disordered silica matrix (Figure 17). Following the discovery of amino acids, DNA was found using molecular techniques by Perry *et al.* (2004) and Kuhlman *et al.* (2006). Also several polymorphic compounds are present in varnishes (Perry *et al.*, 2007) and lipids (Schelbe *et al.*, 2001). These findings suggest that varnish coatings preserve past biology and as discussed later may be an important recorder of contemporaneous life in the local environment.

Silica and varnish

Now, using a battery of techniques including high resolution electron microscopy, recent work has revealed that silica as opal, not clays or metal oxides, is the most important mineral present in desert varnish (Figures 19 and 20). When moisture is available, depressions on rock surfaces form complex chemical cauldrons (Figure 7) in which silicic acid may form leaving varnish structures in the depressions (Figure 8). Evaporation of water

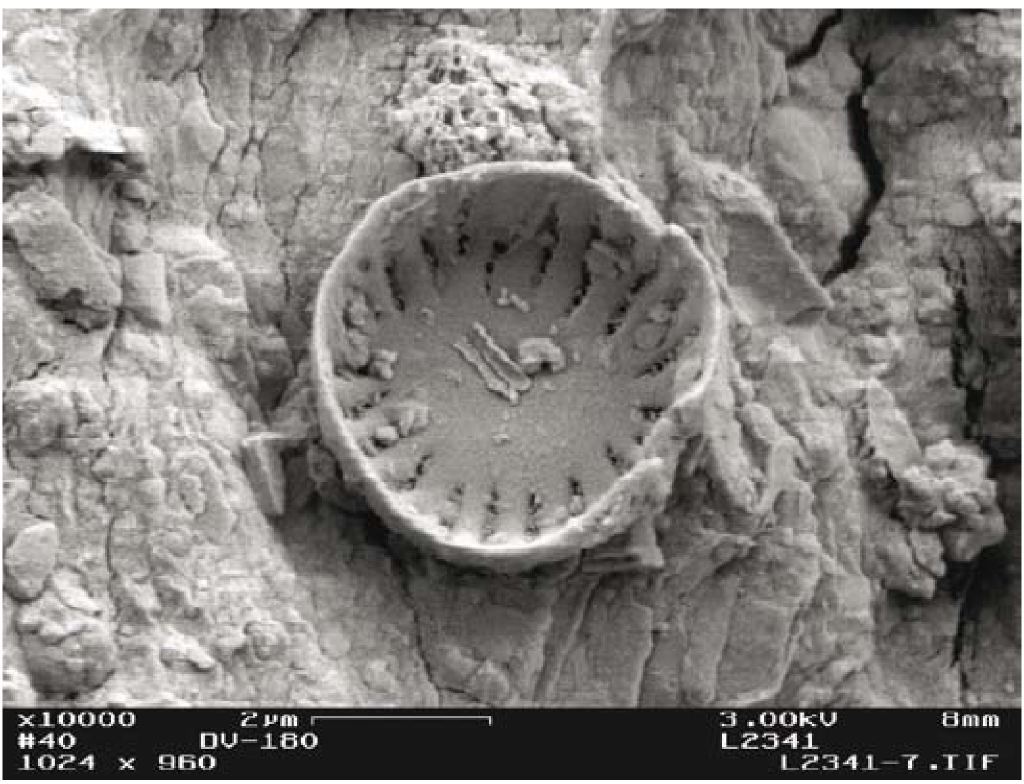

Fig. 15. Diatom on the surface of a desert varnish covered rock. Desert varnish is like sticky paper and whatever lands on the surface may become part of the coating.

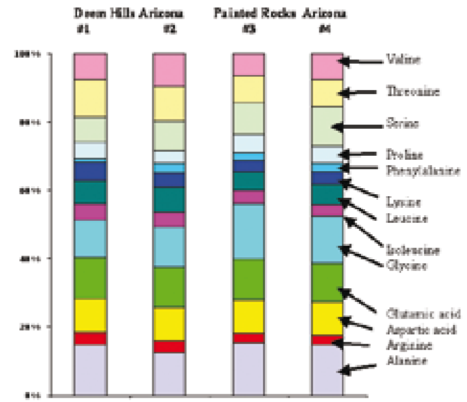

Fig. 16. Relative abundance of 13 protein amino acids found in desert varnish (Perry et al., 2003).

eventually causes concentrations to increase and, eventually, condensation of the silicic acid occurs to produce a gel. During gel formation, surface detrital material and organic compounds

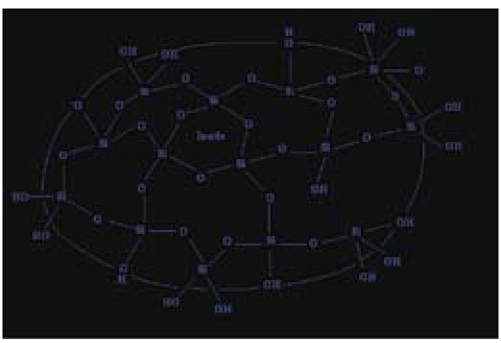

Fig. 17. Amorphous hydrated silica or polymerized silicic acid depicting random bond angles.

are incorporated and when the gels are dried and dehydrated the components are entombed in a lustrous rock coating (Perry *et al.*, 2006, Perry *et al.*, 2007). Evidence of the action of occasional water is forthcoming from analyses which indicate that despite being formed in arid environments, the coatings have been shown to contain up to 9% water (Perry, 1979). This in itself represented a conundrum until the hypothesis of water-rich silica was put forth.

Red coatings on the under surfaces are silica-rich. They do not contain enough manganese to make them black but do have a few weight percent Fe oxide that tints them red. The undercoatings reflect the nature of the mineralogy of the soils they are sitting on. The red undercoating (Figures 5, 21 and 22) is probably formed as water transpires from below and a solution of silicic acid. The soil in this particular case is silica-rich granites. Where stones reside in 'dirt' undercoatings are not usually observed.

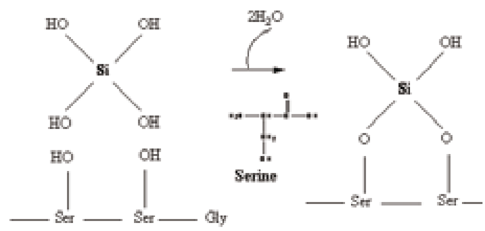

Fig. 18. Possible condensation reaction of silicic acid with hydroxyl groups of serine. Adapted from Zubay, G. Origins of life on the Earth and in the Cosmos (2000) p391.

We can then conclude that silica in desert varnish (Figure 23), red undercoats and silica glazes and even hot-spring silica-rich sinters are made by similar processes (Perry and Lynn, 2006). Desert varnish then can be thought of as a silica glaze enriched in oxides.

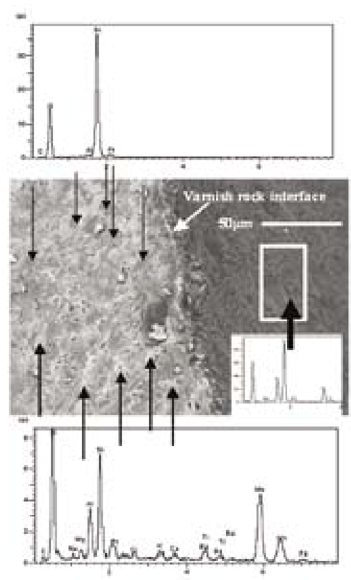

Fig. 19. SEM with EDS, backscatter, and secondary imaging. Lighter elements appear darker and heavier elements appear lighter. Top EDS shows areas that are lighter are composed almost exclusively of Si. Lower EDS shows heavier elements. Lower spectrum is oxide-rich and also has relatively more carbon. The area EDS insert is of the rock substrate and, using electron microprobe, the rock analyses is consistent with phonolite. Adapted from Perry et al., Geology (2006).

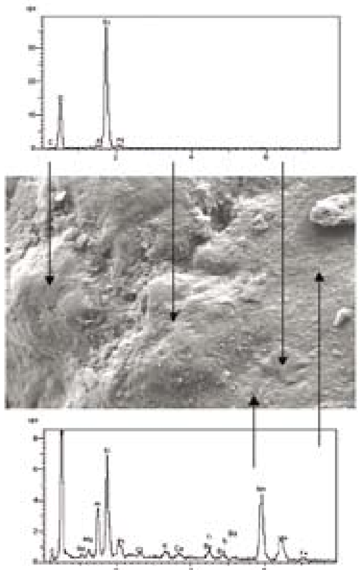

Fig. 20. SEM image of desert varnish surface Baker, California. EDS analyses show silicon-rich mounds ~2-4µm in diameter (top EDS) and oxide enhanced regions (lower EDS). The silicon rich areas (top EDS) have detectable signals only for C, O, Al, other than Si. The Pt signal is for platinum that was used to coat the sample. The spectra vary for the different spots but, the spectra shown are representative for all spots analyzed including the ones depicted by the black arrows. Adapted from Perry et al., Geology (2006).

Hypotheses of formation must stand up to scientific testing and it has often been said that if we could only make a synthetic coating in the laboratory that we must inevitably understand how coatings are made in nature. Figure 10 is an X-ray microprobe map of just such a laboratory generated coating. Laboratory experiments support the role of silica polymerization. Surprisingly it turns out to be a relatively easy process to make coatings in the laboratory (Perry *et al.*, 2005). Conversely, several attempts to make coatings using microbes produced no coatings further supporting non-biological explanations for desert varnish. It cannot be ignored however, that biological organic compounds may effect the chemical formation of coatings when they are present.

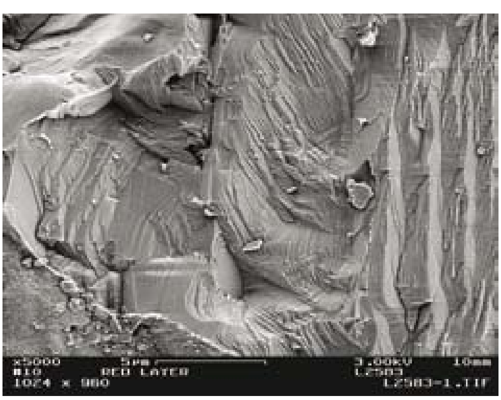

Fig. 21. SEM image of bright red under-glaze (shown in Fig.5). It has a lustrous glossy look under low magnification. However, as the magnification is increased with SEM, the morphology changes dramatically. Figure 22 shows a variety of angular textures with x5,000 magnification. The textural quality is "sintered" and angular but also exhibits a layered morphology.

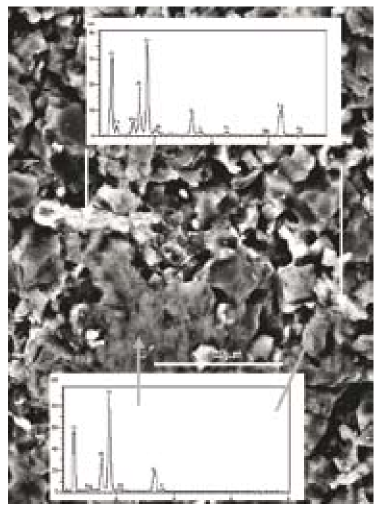

Fig. 22. SEM with EDS of red bottom coat Baker, California, Mojave Desert. Image is a combination of backscatter and secondary electrons. The lighter areas (upper EDS) are heavier elements, the lighter gray areas are lighter elements (lower EDS). Scale bar 20 µm.

STEM with HAADF detectors

SEM with EDAX of a desert varnish coating indicates silicon-rich surface areas (Figures 19-20). High-resolution transmission electron microscopy also produced evidence for amorphous hydrated silica (Figure 23) in powdered samples from the Gobi Desert, the Sonoran Desert, the Mojave Desert, and the Namibian Desert. A wafer (~100 nm thick) was cut using FIB (Figure 24) and investigated using a STEM with a HAADF detector and an EDAX. Silicon-rich areas are identified by atomic values using the HAADF detector and then colour enhancing the images to better illustrate the light and heavy element areas within the FIB waffer (Figures 25-27).

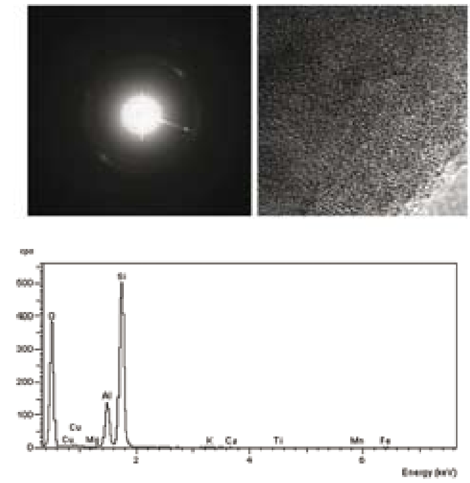

Fig. 23. TEM image of Gobi Desert black varnish coating (upper right). EDS showing primarily Si and Al. Diffraction pattern (upper left) indicates limited order. The high magnification TEM image (upper right) suggests that this coating sample is only partially amorphous.

Desert varnish as a biological recorder

Our work indicates that biology is not required for desert varnish formation and the source of the organic components is from outside rather than within the varnish. Importantly however, the accumulation of environmental products in desert coatings preserves a biological, climatological and environmental record. The record contained in desert varnish extends through time because it occurs in very fine layers that grow over each other. The growth pattern is very similar to that seen in stromatolites (layers sometimes produced by the growth of particle-binding bacteria which ultimately harden to form rock) and each layer records the environment in which it formed. The deepest, oldest layers in the varnish may

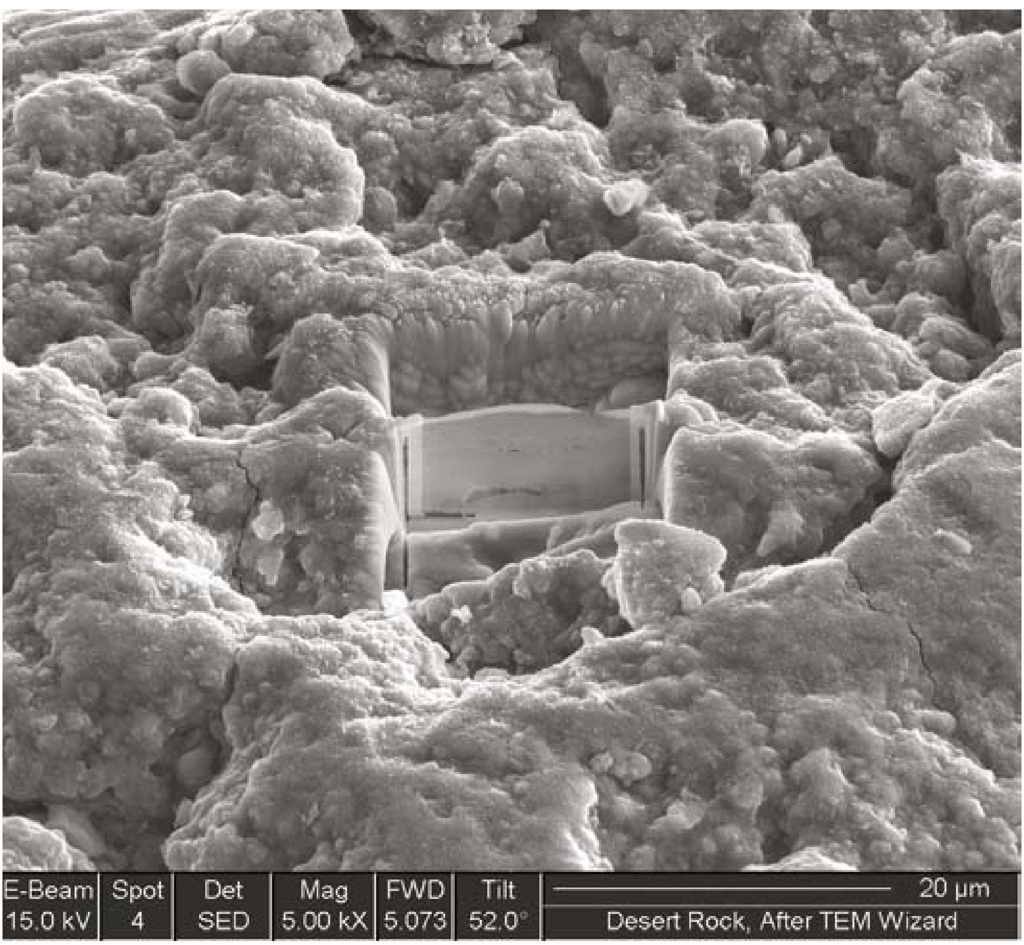

Fig. 24. Surface area of desert varnish showing where a FIB section for STEM analyses was extracted.

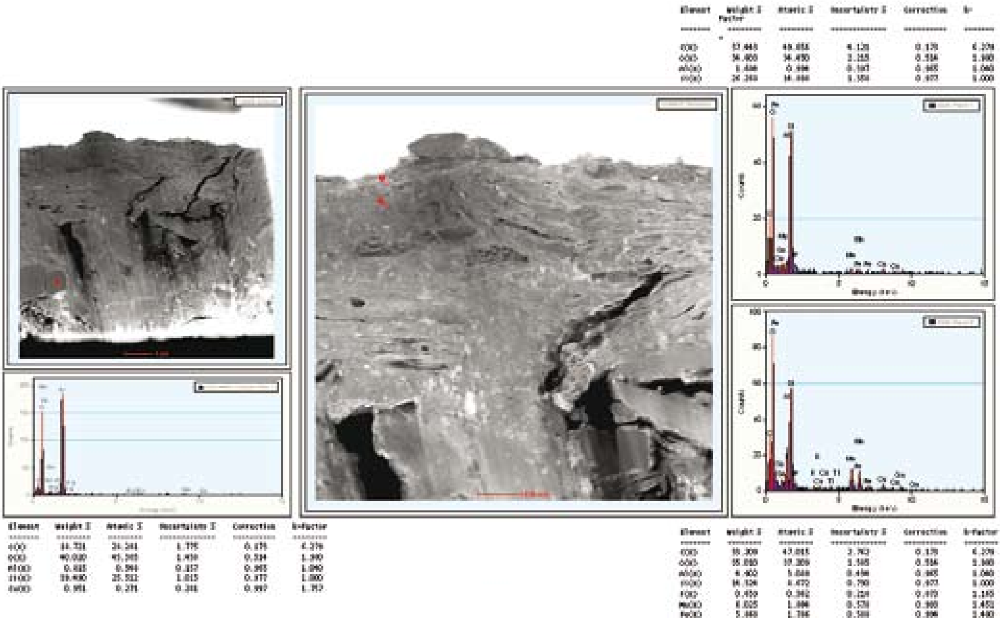

Fig. 25. HAADF STEM images and EDX point analysis. Red scale bar (centre image) is 500 nm.

have formed in very different conditions to the shallowest, youngest layer. These layers, and all the ones in between, represent a record of environmental change. So these lustrous chroniclers of the local surroundings can provide a window back in time.

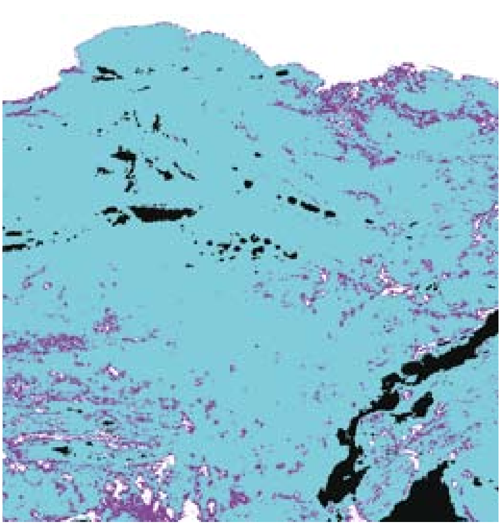

Fig. 26. STEM colour enhanced whereby colours are assigned to a range of z-values from the grey tones imaged by the half detector. Si is represented by cyan. Purple and white represents higher atomic value oxides such as iron and manganese. Black are vacant areas in the FIB section. (See Fig. 25 for scale bar)

Desert varnish and cultural records

On a barren hillside 40 miles east of Fallon, Nevada are darkly stained rock outcrops (Figure 28). The sloping hillside climbs for a few hundred feet above an ancient lakebed. The shoreline of the now dry Lake Lahonton at Grimes Point, where rock surfaces contain rock writing or "petroglyphs". The rock art incised in the dark coatings is of three main types estimated to be from a few thousand years to as much as 10,000 years before present. The oldest of these petroglyphs are thought to be part of ritual ceremonies where shamans incised the rock. Called 'Pit and Groove' style, they resemble small craters (Figure 28). Petroglyphs are incised in varnish coatings all over the world. When varnish coatings lack manganese they appear red (Figure 29). In addition to providing valuable cultural information the petroglyphs also reveal the formation rates of desert varnish. Rock exposed during the making of the ~10,000 year old petroglyph has a new layer of desert varnish while the younger ~2000 year old petroglyph is not re-covered with varnish. This suggests that desert varnish formation occurs over thousands of years rather than hundreds of years at Grimes Pt.

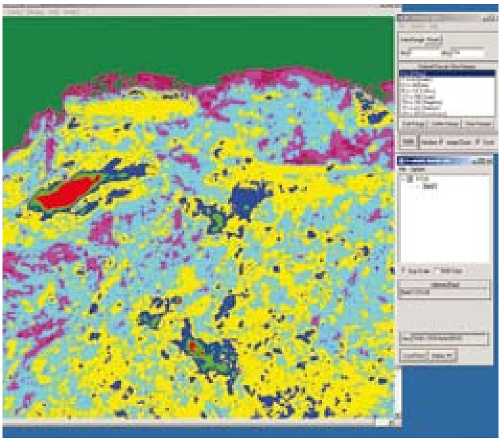

Fig. 27. Showing STEM with z values colour enhance using a remote sensing program. Silica z values are in both the yellow and cyan colour range. Purple are heavier elements such as oxides. (See Fig. 25 for scale bar)

Fig. 28. Petroglyphs at Grimes Point National Monument, Nevada. The insert is of a petroglyph dated by archaeologists at ~2000 years old, while the main photo is of a style of petroglyph thought to be ~10,000 years old. Rock exposed during the making of the ~10,000 year old petroglyph is recovered with varnish while the younger petroglyph is not re-covered with varnish.

Desert varnish on Mars

Recently, camera images from the Mars Pathfinder landing site have strongly suggested the presence

Fig. 29. Petroglyphs from Painted Rocks, Arizona.

of desert varnish on many Martian rocks (Murchie *et al.*, 2004). Angular, equant, and tabular rocks and large boulders display spectral characteristics suggesting varying levels of ferric minerals. These minerals form part of a desert varnish-like coating that may have formed in the presence of thin films of water when Mars had a moister climate than at the present day.

If silica exists in desert varnish-like coatings on Mars, as on Earth, then it may contain chemical signatures of previous life (Perry and Hartmann, 2006). It is possible that on Mars the earliest formed layers in any stromatolite-like sequence may have recorded a wetter more biologically-amenable Martian environment. Moreover it is likely that Martian desert varnish would be a better preserver of organic matter that its Earth counterpart (Perry and Sephton, 2006). The current Martian environment is much colder and drier than that on Earth. Martian desert varnish records may also extend further back in time. On Earth desert varnish is a relatively recent rock coating generated in time periods commonly less than 100,000 years; older examples are removed by physical and chemical weathering. Evidence on Mars of ancient surfaces and events suggest that physical and chemical weathering is less aggressive than on our planet.

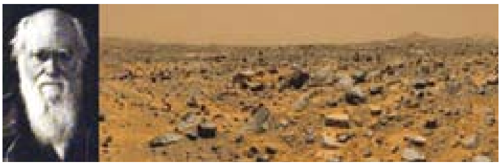

Fig. 30. The cold and stable desert environment on Mars which may be the host for extraterrestrial examples of biochemical desert varnish. Do Mars rocks have a desert varnish coating? Would Darwin wonder if there was desert varnish on Mars? And if so, would they show that there was early biological evolution on Mars?

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