

REVIEW

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Life on Mars: chemical arguments and clues from Martian meteorites

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Abstract Primitive terrestrial life – defined as a chemical system able to transfer its molecular information via self-replication and to evolve – probably originated from the evolution of reduced organic molecules in liquid water. Several sources have been proposed for the prebiotic organic molecules: terrestrial primitive atmosphere (methane or carbon dioxide), deep-sea hydrothermal systems, and extraterrestrial meteoritic and cometary dust grains. The study of carbonaceous chondrites, which contain up to 5% by weight of organic matter, has allowed close examination of the delivery of extraterrestrial organic material. Eight proteinaceous amino acids have been identified in the Murchison meteorite among more than 70 amino acids. Engel reported that L-alanine was surprisingly more abundant than D-alanine in the Murchison meteorite. Cronin also found excesses of L-enantiomers for nonprotein amino acids. A large collection of micrometeorites has been recently extracted from Antarctic old blue ice. In the 50- to 100- μm size range, carbonaceous micrometeorites represent 80% of the samples and contain 2% of carbon, on average. They might have brought more carbon than that involved in the present surficial biomass. The early histories of Mars and Earth clearly show similarities. Liquid water was once stable on the surface of Mars, attesting the presence of an atmosphere capable of decelerating C-rich micrometeorites. Therefore, primitive life may have developed on Mars as well and fossilized microorganisms may still be present in the near subsurface. The Viking missions to Mars in 1976 did not find evidence of either contemporary or past life, but the mass spectrometer on the lander aeroshell determined the atmospheric composition, which

has allowed a family of meteorites to be identified as Martian. Although these samples are essentially volcanic in origin, it has been recognized that some of them contain carbonate inclusions and even veins that have a carbon isotopic composition indicative of an origin from Martian atmospheric carbon dioxide. The oxygen isotopic composition of these carbonate deposits allows calculation of the temperature regime existing during formation from a fluid that dissolved the carbon dioxide. As the composition of the fluid is unknown, only a temperature range can be estimated, but this falls between 0° and 90°C, which would seem entirely appropriate for life processes. It was such carbonate veins that were found to host putative microfossils. Irrespective of the existence of features that could be considered to be fossils, carbonate-rich portions of Martian meteorites tend to have material, at more than 1000 ppm, that combusts at a low temperature; i.e., it is an organic form of carbon. Unfortunately, this organic matter does not have a diagnostic isotopic signature so it cannot be unambiguously said to be indigenous to the samples. However, many circumstantial arguments can be made to the effect that it is cogenetic with the carbonate and hence Martian. If it could be proved that the organic matter was preterrestrial, then the isotopic fractionation between it and the carbon is in the right sense for a biological origin.

Key words carbonates · Isotopes · Mars · Martian meteorites · Micrometeorites · Nanofossils · Prebiotic chemistry

Origin of life on the primitive Earth

Primitive life probably appeared with the first chemical systems able to transfer their molecular information via self-replication and to evolve in liquid water. Unfortunately, the direct clues that may help chemists to identify the molecules which participated in the emergence of life on Earth about 4 billion years (4Ga) ago have been erased by plate tectonics, the permanent presence of running water, unshielded solar ultraviolet radiation, and oxygen produced by life. By

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analogy with contemporary life, it is generally believed that primitive life originated from the processing of reduced organic molecules by liquid water. Primitive life may also have begun based on mineral crystals. This idea has been developed by Cairns-Smith (1982) but still needs to be supported by experiments.

Liquid water

It is generally thought that life on Earth emerged in liquid water. For this reason, water is considered as one of the prerequisites for life to appear and evolve on a planet. In contemporary living systems, water participates as a solvent in the organization of biopolymers in three-dimensional structures and as a chemical partner in most of the biochemical pathways. According to its molecular weight, water should be a gas under standard terrestrial conditions by comparison with CO₂, SO₂, H₂S, etc. Its liquid state is the result of its ability to form hydrogen bonds. This characteristic is not restricted to water molecules, because alcohols exhibit a similar behavior. However, the polymeric network of water molecules via H-bonds is so tight that the boiling point of water molecules is raised from 40°C (temperature inferred from the boiling point of the smallest alcohols) to 100°C. Hydrogen bonds are formed between water molecules and organic molecules if the latter contain –OH, –NH, or –SH groups in addition to carbon and hydrogen. As a consequence of this affinity, many CHONS organic molecules are soluble in water. Hydrocarbons cannot form H-bonds with water; these molecules are not soluble in water and escape from water as much as possible. In addition to the H-bonding capability, water exhibits a large dipole moment (1.85 debye) as compared to alcohols (<1.70 debye). This large dipole moment favors the dissociation of ionizable groups such as –NH₂ and –COOH, leading to ionic groups that can form additional H-bonds with water molecules, thus improving the solubilization. Water is also an outstanding dielectric. When oppositely charged groups are formed, their recombination is not favored because the attraction force for reassociation is proportional to 1/ε. Abiotic organic molecules can be divided into two families: hydrocarbons and CHONS-containing molecules. When brought into the presence of liquid water, hydrocarbons try to escape the water molecules whereas CHONS, especially those that bear ionizable groups, have some affinity for water. When these two species are forced to coexist within the same molecules, the duality generates interesting topologies. When they are separated by long distances, such as in fatty acids or phospholipids, then micelles, vesicles, or liposomes are formed from the clustering of the hydrophobic groups. Over short distances, hydrophobic and ionizable groups generate chain conformations that depend strongly on the sequence, i.e., the distribution of the group along the chains.

Water molecules are widespread in the universe as grains of solid ice or as very dilute water vapor. Liquid water is a fleeting substance that can persist only above 0°C and at an

atmospheric pressure greater than 6 mbars. Therefore, the size of a planet and its distance from the star are two basic characteristics that determine the presence of liquid water. If a planet is too small, like Mercury or the Moon, it will not be able to retain any atmosphere and, therefore, liquid water. If the planet is too close to the star, the mean temperature rises because of starlight intensity. Any seawater present would evaporate, delivering large amounts of water vapor to the atmosphere and thus contributing to the greenhouse effect. Such a positive feedback loop could lead to a runaway greenhouse: all the surface water would be transferred to the upper atmosphere where photodissociation by ultraviolet light would break the molecules into hydrogen, which escapes into space, and oxygen, that combines with the crust. If a planet is far from the star, it may host liquid water providing that it can maintain a constant greenhouse atmosphere. However, water would provoke its own disappearance. The atmospheric greenhouse gas, CO₂, for instance, would be dissolved in the oceans and finally trapped as insoluble carbonates by rock weathering. This negative feedback would lower the surface pressure and temperature to an extent that water would be largely frozen. The size of the Earth and its distance from the sun are such that the planet never experienced either a runaway greenhouse or a divergent glaciation.

Terrestrial production of reduced organic molecules

Oparin (1924) suggested that the small reduced organic molecules needed for primitive life were formed in a primitive atmosphere dominated by methane. The idea was tested in the laboratory by Miller (1953) when he exposed a mixture of methane, ammonia, hydrogen, and water to electric discharges. In his initial experiment, he obtained 4 of the 20 naturally occurring amino acids via the intermediary formation of hydrogen cyanide and formaldehyde. Miller's laboratory synthesis of amino acids occurs efficiently when a reducing gas mixture containing significant amounts of hydrogen is used. However, the true composition of primitive Earth's atmosphere remains unknown. Today, geochemists favor a neutral or weakly reducing atmosphere dominated by carbon dioxide (Kasting 1993). Under such conditions, the production of amino acids appears to be very limited (Schlesinger and Miller 1983).

More recently, Wächtershäuser (1994) suggested that the carbon source for life was carbon dioxide. The energy source required to reduce carbon dioxide was provided by the oxidative formation of pyrite (FeS₂) from troilite (FeS) and hydrogen sulfide. Pyrite has positive surface charges and bonds the products of carbon dioxide reduction, giving rise to a two-dimensional reaction system, a surface metabolism. Experiments are presently being run to verify this new hypothesis. Deep-sea hydrothermal systems may also represent likely environments for the synthesis of prebiotic organic molecules. Experiments have been carried out to test whether amino acids can be formed under conditions

simulating the hydrothermal vents (Holm and Andersson 1995).

Extraterrestrial delivery of organic molecules to the Earth

Comets show substantial organic material. According to Delsemme's analysis, comet Halley dust particles ejected from the nucleus contain 14% organic carbon by mass (Delsemme 1991). About 30% of cometary grains are dominated by light elements, C, H, O, and N, and 35% are close in composition to carbonaceous chondrites (Kissel and Krueger 1987; Langevin et al. 1987). Among the molecules identified in comets are hydrogen cyanide and formaldehyde. The presence of purines, pyrimidines, and formaldehyde polymers has also been inferred from the fragments analyzed by Giotto Picca and Vega Puma mass spectrometers. However, there is no direct identification of the complex organic molecules present in the dust grains and in the cometary nucleus. Many chemical species of interest for exobiology were detected in comet Hyakutake in 1996, including ammonia, methane, acetylene (C₂H₂), acetonitrile (CH₃CN), and hydrogen isocyanide (HN=C) (Hearn [in press]; Irvine et al. 1996). In addition to hydrogen cyanide and formaldehyde (H₂CO), seen in several earlier comets, the comet Hale-Bopp was also shown to contain methane, acetylene, formic acid (HCOOH), acetonitrile, hydrogen isocyanide, isocyanic acid (HNCO), cyanoacetylene H-C≡C-CN, and thioformaldehyde (H₂C=S). Cometary grains may have been an important source of organic molecules delivered to the primitive Earth (Oro 1961; Chyba et al. 1990; Delsemme 1992; Greenberg 1993).

The study of meteorites, particularly the carbonaceous chondrites that contain up to 5% by weight of organic matter, has allowed close examination of extraterrestrial organic material delivered to the Earth. Eight proteinaceous amino acids have been identified in the Murchison meteorite among more than 70 amino acids (Cronin et al. 1988). These amino acids are asymmetric and the two-handedness, the L and D forms, are generally found in equal proportions. Engel reported that L-alanine and L-glutamic acid were surprisingly more abundant than the corresponding D-enantiomers (L-enantiomer excesses of more than 30% and 50%, respectively) in the Murchison meteorite (Engel and Macko 1997). Cronin found also excesses of L-enantiomers for nonprotein amino acids in the same meteorite (Cronin and Pizzarello 1997). The presence of L-enantiomeric excesses in the Murchison meteorite suggests an extraterrestrial asymmetric synthesis of amino acids, asymmetry that is preserved inside the meteorite. These excesses found in the Murchison meteorite may help to understand the emergence of an one-handed (homochiral) primitive life. Indeed, homochirality is now believed to be not just a consequence of life, but also a prerequisite for life, because stereoregular structures such as protein β-sheets do not form with mixtures of amino

acids of both orientations. The excess of one-handed amino acids found in the Murchison meteorite may result from the processing of the organic mantles of interstellar grains by circularly polarized synchrotron radiation from a neutron star remnant of a supernova (Bonner and Rubenstein 1987). Recent dust collection, both above terrestrial atmosphere by Brownlee (Love and Brownlee 1993), and in the Greenland and Antarctica ice sheet by Maurette et al. (1995), show that the Earth captures interplanetary dust as micrometeorites at a rate of about 50–100 tons per day. About 99% of this mass is carried by micrometeorites in the 50- to 500-μm size range. This value is about 2000 times higher than the most reliable estimate of the meteorite flux (about 0.03 tons per day) recently estimated by Bland et al. (1996).

This amazing dominance of micrometeorites already suggests their possible role in delivering complex organics to the early Earth 4.2–3.9 billion years (Ga) ago when the micrometeorite flux was enhanced by a factor of 1000 (Anders 1989). In the Antarctic micrometeorites, a high percentage of unmelted chondritic micrometeorites from 50 to 100 μm in size has been observed, indicating that a large fraction crossed the terrestrial atmosphere without drastic thermal treatment. In this size range, the carbonaceous micrometeorites represent 80% of the samples and contain 2% of carbon, on average. They might have brought about 10²⁰ g of carbon over a period of 300 million years, corresponding to the late terrestrial bombardment phase. This delivery represents more carbon than that engaged in the surficial biomass, i.e., about 10¹⁸ g. Amino acids such as α-amino isobutyric were identified in these Antarctic micrometeorites. These grains contain also a high proportion of metallic sulfides, oxides, and clay minerals that belong to various classes of catalysts. Thus, in addition to the carbonaceous matter, the micrometeorites may also have delivered a rich variety of catalysts, having perhaps acquired specific crystallographic properties during their synthesis in the microgravity environment of the early solar nebula. They may have functioned as tiny chondritic chemical reactors when reaching oceanic water.

Was Mars similar to the Earth?

The early histories of Mars and Earth clearly show similarities. Geological observations collected from Martian orbiters suggest that liquid water was once stable on the Mars surface, attesting to the presence of an atmosphere capable of decelerating carbonaceous micrometeorites. Mars mapping by Mariner 9 and by Viking 1 and 2 revealed channels resembling dry river beds. Three major classes of channels were identified: (i) dendritic runoff channels and valleys primarily associated with cratered terrain older than 3.8 Ga and which are generally associated with fluvial water erosion; (ii) outflow channels suggestive of large-volume flows in cataclysmic events; and (iii) fretted channels that are steep walled with smooth flat floors suggestive of erosion by debris flows. The total amount of water that may have

existed at the surface of Mars is difficult to estimate and varies from some meters to several hundred meters. The climatic conditions required for the formation of the valley networks are also poorly understood. The most plausible explanation is a relatively thick CO₂ atmosphere (1–5 bars) in the early history of Mars, giving rise to a greenhouse effect that would have allowed for mean global surface temperature to remain above the freezing point of water. It has recently been suggested (Carr 1995) that the valley networks, like the fretted channels, formed mainly by mass wasting, aided by groundwater seepage into the mass-wasted debris, between 3.8 and 3.7 Ga ago. Therefore, liquid water seems to have been restricted to the very early stages of Martian history. By about 3.8 Ga ago, atmospheric CO₂ was probably irreversibly lost because of carbonate formation and pressure and declining temperature.

The Viking 1 and 2 lander missions were designed to address the question of extant life on Mars. Three experiments were selected to detect metabolic activity such as photosynthesis, nutrition, and respiration of potential microbial soil communities. The results were ambiguous, because although “positive” results were obtained no organic carbon was found in the Martian soil by gas chromatography-mass spectrometry. It was concluded that the most plausible explanation for these results was the presence, at the Martian surface, of highly reactive oxidants such as H₂O₂ that would have been photochemically produced in the atmosphere (Hartman and McKay 1995). The Viking lander could not sample soils below 6 cm and therefore the depth of this apparently organic-free and oxidizing layer is unknown. Bullock et al. (1994) have calculated that the depth of diffusion for H₂O₂ is less than 3 m. Direct photolytic processes could also be responsible for the dearth of organics at the Martian surface (Stoker and Bullock 1997).

The alpha-proton X-ray spectrometer (APXS) on board the rover of the Mars Pathfinder mission measured, in 1997, the chemical composition of six soils and five rocks at the Ares Vallis landing site. The analyzed rocks were partially covered by dust or a weathering rind similar in composition to the dust. Some rocks are similar to terrestrial andesites, but it is not certain that these rocks are igneous. The texture of other rocks is difficult to interpret and might be sedimentary or metamorphic (Rieder et al. 1997).

Martian meteorites

Although the Viking missions were disappointing in the first instance for exobiology, in the long run the program has proven extremely beneficial for the investigation of the possibility of life on Mars. Before Viking, it had been apparent that there was a small group of meteorites, all of igneous origin, called the SNC (after their type specimens: Shergotty, Nakhla, and Chassigny), that had comparatively young crystallization ages equal to or less than 1.3 Ga (Jagoutz and Wänke 1986). In 1979 another one of these meteorites was found on Antarctica. This specimen, EETA

79001, had trapped gas within glass pockets that both compositionally and isotopically matched, in all respects, the makeup of the Martian atmosphere as measured by the mass spectrometer utilized for assessing the soil for the presence of organic compounds (Bogard and Johnson 1983; Becker and Repin 1984; Carr et al. 1985). The data provide a very strong argument that at least that particular SNC meteorite comes from Mars, thus supporting the circumstantial conclusion that materials of young age must have derived from a planetary-sized body. There are now 12 SNC meteorites known in total; others were found more recently on the Antarctic Ice Cap, including one of much older age, ALH 84001. They can all be shown to be related by comparing their oxygen isotopic compositions. Only these 12 specimens (of a total of ~20 000 meteorites) define a correlation line of slope 0.5 on a plot of $\delta^{18}\text{O}$ vs. $\delta^{17}\text{O}$ ¹, with $\Delta^{17}\text{O} = 0.321 \pm 0.013\%$, i.e., displaced from the Earth reference line.

The presence of carbonates in Martian meteorites

The discovery of trapped atmospheric compounds in EETA 79001, particularly as the Martian atmosphere is more than 95% carbon dioxide, had very exciting repercussions for understanding the planet's environment. A knowledge of the carbon isotopic composition of the CO₂ allowed studies seeking the existence of carbonate to proceed. Geochemical procedures involving acid dissolution and stepped combustion pyrolysis very quickly indicated that carbonate minerals were indeed present in small quantities, but it was not for some time that their existence was petrologically confirmed.

Measurement of the three isotopic composition of oxygen would unambiguously establish that the carbonates were Martian; unfortunately, at the present time it is impossible to perform such an analysis with sufficient precision. Therefore there has been a lengthy debate as to whether the carbonates were terrestrial contamination in rocks that were in all other respects igneous (and hence should not contain carbonates). The main problem is that some of the carbonate contains live ¹⁴C, which must be of terrestrial origin (Wright et al. 1997a). The recognition that ALH 84001 was a Martian rock – this was misassigned as another kind of meteorite, a diogenite, for a number of years (Mittelfehldt 1994) – was very important because the carbon isotopic composition of carbonate was measured at $\delta^{13}\text{C} > 40\%$, which is far removed from any terrestrially known carbonate (Romanek et al. 1994). There it is impossible to argue that the vast majority of carbonate is any other than extraterrestrial.

At least two other Martian meteorites have carbonate with elevated $\delta^{13}\text{C}$ values (Carr et al. 1985; Wright et al.

¹ In stable isotope research compositions are discussed as differences (δ values) from internationally agreed standards, given in parts per thousand (‰); positive values denote an enrichment in the minor heavier isotope, negative values a depletion. $\Delta^{17}\text{O}$ is the gross difference from the bulk Earth composition

1989), and amounts of the mineral it would be difficult to assign to terrestrial sources, especially for Nakhla, which was an observed fall. ALH 84001 has many veins and cracks that are lined with rosettes of orange iron/magnesium-containing carbonate minerals (Mittelfehldt 1994; Romanek et al. 1994). The texture of the rock has every appearance that it was inundated by a hydrothermal fluid: water with dissolved carbon dioxide.

Formation conditions of Martian carbonates

If the concentration of CO₂ within the water percolating through Martian meteorites were known, a precise temperature for the deposition of the carbonate could be calculated from the δ¹⁸O composition (Romanek et al. 1994). Without this information only boundary conditions can be set, and the limits appear to be 0° and 90°C for ALH 84001. The higher the δ¹⁸O, the lower the temperature at which deposition occurred. Both the other meteorites that contain significant amounts of carbonate, EETA 79001 and Nakhla, in fact have carbonates with higher δ¹⁸O values than ALH 84001. There are other techniques such as chemical composition for estimating the temperature at which carbonate was formed; some of these give very high temperatures, up to 700°C (Harvey and McSween 1996). However, it should be borne in mind the low temperatures referred to earlier were derived for the whole of the carbonate by reacting with phosphoric acid to give carbon dioxide. The high temperature estimates have predominantly been obtained using chemical analysis methods employing focused probe techniques.

The time of carbonate formation

Assuming carbonate production in Martian meteorites occurred at low temperature, then such a model argues for the existence of liquid water on Mars long after the time implied from photography of surface features, i.e., 3.5 Ga. It is extremely difficult to date carbonates using radioactive decay. Various methods have been applied to the carbonates of ALH 84001; the interpretation of the results is again model dependent. One careful study suggests an age of about 1.3 Ga (Lugmair and Wadhwa 1996), which is interesting because it coincides with an apparent time of the resetting of radioactive clocks on Mars as indicated by the formation dates of various meteorites. Of course, the carbonate in Nakhla cannot be older than 1.3 Ga because this is the formation age of that meteorite. EETA 79001 is even more intriguing in this respect because it has a crystallization age of only 0.18 Ga and was on the surface of the planet until it was ejected 600 000 years ago, according to cosmic ray exposure dating. Water could therefore have been present on Mars in discrete situations until comparatively quite recently. Incidentally, hydrated minerals of clay-type structure have been known in Nakhla for more than two decades (Hutchison 1975).

Organic matter in Martian meteorites

All three of the samples that have carbonates with elevated δ¹³C have coexisting organic matter (Wright et al. 1989; Grady et al. 1994). In EETA 79001, the best studied, samples of a variety of lithologies have been investigated. Without exception specimens without carbonate have a uniformly low organic matter content, as estimated by the ignition temperature established during stepped combustion experiments. In contrast, for the carbonate-rich fractionations, including some extracted from completely glass-sealed inclusions, organic contents are significant. The amounts of carbon are a factor of 5 higher than any other bulk Martian meteorite, although some of these are known to have long been in museum collections and heavily contaminated. The implication is that the organic matter associated with carbonates came to Earth with the meteorite and is cogenetic.

This matter is extremely controversial, and a number of investigators have attempted to argue that the organic material is terrestrial contamination. Hypotheses concerning how it penetrated the meteorite have been put forward; some of these are highly specific, such as penetration of the fabric of the meteorite by Antarctic meltwater containing polyaromatic hydrocarbons (PAHs). The advocates of such ideas do not seem to recognize the degree to which the passage of terrestrial water would disturb the oxygen isotopic systematics of the silicates. None of the Antarctica Martian meteorites show evidence for major oxygen isotope exchange while on Earth, which makes the Martian Δ¹⁷O line much more difficult to constrain.

Unfortunately, none of the organic matter in EETA 79001, or for that matter in Nakhla and ALH 84001 with the exception of minute amounts of PAHs in the latter (Mackay et al. 1996), has been fully characterized in terms of compound class or individual structures. This is a wide-open field but one requiring a very painstaking, high-sensitivity approach. The structure of molecules within this fraction might be expected to be diagnostic of its origin.

Isotopic considerations

Some indication of origins might be forthcoming from the fractionation of carbon isotopes between carbonate and organic phases (Wright et al. 1997b). The shift seen in the meteorites is large and thus predicts a low-temperature event. On Earth, Schidlowski and his co-workers (1987) have compiled the carbon isotopic composition of more than 1600 samples of fossil kerogen (a complex organic macromolecule produced from the debris of biological matter) with carbonates in the same sedimentary rocks. The organic matter has an average δ¹³C of -25‰ ± 7‰, whereas the carbonate varies by no more than about 2‰ around a mean of 0‰. The approximate 25%–30% shift is taken as an indication that biosynthesis by photosynthetic organisms was involved; in fact this offset is now taken to be

one of the most powerful indications that life on Earth was active nearly 3.9 Ga ago because the sample suite encompasses specimens right across the geological time scale. Some organic matter in ancient sediments has been measured as being even more enriched in the light isotope of carbon, which would suggest involvement of methane-utilizing organisms. The difference between organic matter and carbonates in Martian meteorites is greater than the shift seen on Earth, which could also be indicative of biosynthesis, but some as yet unknown reason cannot be ruled out.

Nanofossils in ALH 84001

By far the most controversial observation made concerning Martian meteorites is the suggestion that organized nanometer-sized structures have been seen in the regions of ALH 84001, showing evidence for aqueous activity and carbonate deposition. Based on the similarity of these forms with filamentous living organisms and microfossils in terrestrial sediments, the Martian features have also been proposed as fossils (Mackay et al. 1996). These observations and conclusions have been the subject of intense media interest. Although the enthusiasm with which they have been received in some quarters may have been misplaced, on the basis of the principle of Occam's razor (the simplest explanation is best), the finding of simple structures in a region in which there is evidence that water passed leaving both carbonate minerals and organic detritus might be the best evidence we have yet for the existence of past life on our sister planet. This is however a tantalizing observation that needs much more evidence for confirmation.

The study of all these features in Martian meteorites, and the discovery of whether they are related or disparate, is a fascinating challenge. Whatever their underlying cause, it does already appear that suppositions that Mars was totally inhospitable to life were premature, and the future exploration of the planet will address this and related questions.

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