



Review A Physicochemical Consideration of Prebiotic Microenvironments for Self-Assembly and Prebiotic Chemistry

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Abstract: The origin of life on Earth required myriads of chemical and physical processes. These include the formation of the planet and its geological structures, the formation of the first primitive chemicals, reaction, and assembly of these primitive chemicals to form more complex or functional products and assemblies, and finally the formation of the first cells (or protocells) on early Earth, which eventually evolved into modern cells. Each of these processes presumably occurred within specific prebiotic reaction environments, which could have been diverse in physical and chemical properties. While there are resources that describe prebiotically plausible environments or nutrient availability, here, we attempt to aggregate the literature for the various physicochemical properties of different prebiotic reaction microenvironments on early Earth. We introduce a handful of properties that can be quantified through physical or chemical techniques. The values for these physicochemical properties, if they are known, are then presented for each reaction environment, giving the reader a sense of the environmental variability of such properties. Such a resource may be useful for prebiotic chemists to understand the range of conditions in each reaction environment, or to select the medium most applicable for their targeted reaction of interest for exploratory studies.

Keywords: origin of life; chemical evolution; reaction microenvironments; physical chemistry; geochemistry

1. Introduction

The early Earth was like a laboratory but without an intervening chemist. That is to say, early Earth possessed a variety of chemicals, reaction vessels/compartments, and conditions [1], generating complex chemical systems without a target, but which managed to self-organize into life. For many of these scenarios, interdisciplinary studies have been carried out to qualify and quantify their prebiotic plausibility. For example, the origins of life community have considered different geological settings (i.e., hot springs or oceans) as plausible "reaction vessels" on early Earth [2–5]. However, these geological settings, while informative about constraining the potential reaction environments, conditions, and chemicals, are mostly still at large length scales of centimeters, meters, or kilometers. Less attention has been paid to early Earth environments at the microscopic level, the length scales that have the potential to directly affect the dynamics of chemical reactions, self-assembly, and cellular/protocellular processes.

Indeed, within larger geological settings, a variety of physical and chemical environments exist at smaller length scales. These smaller 'microenvironments', which are as small as the microliter or micron scale, could vary significantly in physical and chemical properties. Examples include the widely varying temperature and pH conditions in hot springs [6]



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or the simultaneous existence of both aqueous environments as well as supercritical liquid carbon dioxide (CO_2) in the deep ocean [7].

Here, we consider a number of prebiotic microenvironments on Earth and discuss their physical and chemical characteristics and subsequent impact on primitive reactions and/or self-assembly. The intention is to create a resource that researchers can use to guide their laboratory reactions toward more realistic geological conditions or to use specific reaction microenvironments to constrain what prebiotic processes can take place. This review focuses on condensed phase reactions including but not limited to aqueous phase chemistry, interfacial chemistry, and hydrothermal/geochemical synthesis (Figure 1) [1]. We refer the reader to reviews of gas-phase chemistries and in particular, gas-phase photochemistry [8–10].



Figure 1. A variety of chemical processes could have occurred on early Earth. Prebiotic reactions could have occurred in the gas phase (atmospheric synthesis), the aqueous phase, or on material interfaces, just to name a few. Other reactions could have occurred extraterrestrially, followed by delivery to Earth; in this review, we particularly focus on condensed phase reactions on Earth. Figure adapted and reprinted with permission from [9] under a Creative Commons license.

The scope of this paper is primarily on environments generated purely from and/or residing in geological formations, although a number of potential environments generated from self-assembled primitive molecules (such as in the interior of vesicle bilayers [11] or within phase-separated polymer droplets [12,13]) are briefly mentioned. Although a number of reaction environments exist extraterrestrially such as methane surface lakes or ammonium-rich sub-surface lakes on Titan [14], we restrict the environments covered here to only those thought to have occurred on Earth. In the future, we hope to expand this analysis to more extraterrestrial environments as more concrete information on the chemical and physical properties of such environments becomes known, following additional planetary science studies and missions such as the Titan Dragonfly mission [15]. We refer the interested reader to

papers discussing prebiotic chemistry in extraterrestrial environments such as meteorites [16], interstellar ice [17] or gas [18], or other planetary bodies [2,19–21].

2. Prebiotic Microenvironments and Where to Find Them

The goal in this section is to take a physicochemical perspective and categorize specific reaction microenvironments available on early Earth, which may be found within a number of (macroscopic) geological environments and scenarios. The overview of the physicochemical properties is expected to help point to opportunities that different environments present. For completeness, we encourage the reader to consider how the geological context is inextricable from prebiotic chemistry by reading the excellent works on the topic (e.g., [1,22–25]).

We start our overview with bulk polar and non-polar environments. Within these broad categories, different colloidal structures are discussed. Finally, we end with a brief discussion of molten and solid rocks.

2.1. Aqueous Environments

Living processes depend on water. One reason is that liquid water is important for diffusion, thus allowing reactants to meet. Water also hosts acid and base chemistries, and is a polar protic solvent that can readily solubilize ionic compounds and hydrogen bond with solutes. Bulk water's highly polar environment is responsible for the hydrophobic effect [26], liquid–liquid phase separation [27], and promotes amphiphile self-assembly into micelles, membranes, or other structures [28].

On early Earth, just as in the present day, there would have been sources of fresh water including streams, ponds, lakes, hot springs as well as salt water such as oceans and hydrothermal systems containing brines. The range of different salinities is covered in Section 3.1.

2.1.1. Bulk Aqueous Solution

In bulk systems there are still microenvironments that should be taken into account. One important factor to consider is the temporal stability of the aqueous environment. Surficial systems can be subjected to wet/dry or freeze/thaw cycles, and while this is not applicable to larger bodies of water, turbulent mixing still creates temporal instability in any aqueous system where the length scales are larger than the Reynolds number. That said, ocean stratification can broadly lead to different zones of stable salt/density and temperature, with other variables such as nutrient content potentially varying during mixing [29]. If the relevant length scales are small and the velocities are slow such as in rock pores or narrow channels [30], the flow could easily be laminar, and other types of transport processes such as thermophoresis [30,31] or diffusiophoresis [32] could begin to dominate.

2.1.2. Sea Spray (Aqueous Aerosols)

Another form of liquid aqueous environments is in aerosol droplets [33], which could have formed on early Earth due to turbulent waves or wind acting on different bodies of water. While the volumes of water in aerosols are small, the high surface area to volume ratio and ample exposure to sunlight means that surface-based processes and photochemistry can generate new molecules potentially not synthesizable in other environments [33,34] affording, for example, cross-linked lipids [35].

2.1.3. Gels and Other Hygroscopic Environments

Aqueous environments can also exist in forms that are intermediates to the solid and liquid phases. Siliceous hot-spring deposits, which likely would have been present on early Earth, contain hydrophilic amorphous silica that can retain water [36]. Mixtures of organic molecules likely present on early Earth have also been shown to form gels [37,38]. Hygroscopic salts also sequester water [39]. These environments enable unique aqueous-

phase chemistries such as the synthesis of polymer-supported zinc sulfide nanocrystals [40] or photochemical phosphorylation [38], and can also reduce evaporation rates and prevent total desiccation.

2.1.4. Ice

The final form of an aqueous-derived environment considered here is ice, which could have been derived from the freezing of liquid aqueous solutions in different bodies of water on early Earth. While carbon-cycle modeling reveals that early Earth is thought to have been temperate [4], fluctuations in conditions could have potentially created sub-zero temperatures and thus ice. Eutectic phases in ice provide aqueous environments that are concentrated in solutes, leading to reactions that are unfavorable in a dilute aqueous environment; we cover eutectic phases in Section 2.2.1.

2.2. Alternative Liquid Environments

2.2.1. Non-Aqueous Solvents

Aside from water, other liquid environments on early Earth could have been in the form of non-aqueous solvents. Many reactions in prebiotic chemistry are formally known as condensations, which are reactions that covalently join together two compounds while eliminating a molecule of water in the process. The polymerization of amino acids and ribonucleotides into peptides and RNA, respectively, are examples of condensation reactions. In water, these condensations tend not to be spontaneous, partly as a consequence of Le Chatelier's principle, since water as a solvent is present in large excess and pushes the equilibrium towards the reactants. Hence, non-aqueous solvents (i.e., those based on organic compounds) have the potential to make condensation reactions more favorable.

While the modern synthetic organic laboratory has a large variety of non-aqueous (organic) solvents at its disposal, the majority would almost certainly not have been abundantly available on early Earth. One reason is simply the lack of prebiotically plausible synthetic pathways to achieve reservoir amounts of these organic compounds necessary to act as solvents, many of the chemical structures of which can be relatively complex. Another reason is that the temperature and pressure conditions of early Earth limit what potential solvents could have accumulated, even if endogenous prebiotic synthetic pathways were producing them in large quantities. The boiling points could be too low to exist as liquids at room temperature or be significantly lower than that of water so that their concentration from aqueous solutions is not realistic. Only organic liquids that have a higher boiling point than water could have accumulated to excess amounts, a circumstance required for a compound to act as a solvent. With these constraints in mind, it is conceivable, however, that some organic liquids could have accumulated in relatively large excess.

For example, an organic solvent that may have accumulated in certain early Earth geological scenarios is formamide. A formal hydration product of hydrogen cyanide (molecular formula: HCONH₂), formamide has a boiling point of 210 °C under standard pressure and has limited azeotropic associations with water [41]. Formamide could have been produced through multiple pathways [42] including mechanisms that involve atmospheric spark-discharge [43], ionizing radiation such as proton irradiation [44], UV irradiation [45], pyrolysis [46], or thermal reactions promoted by catalysis [47]. While it is unclear whether very large pools of formamide could have existed on early Earth (at least in comparison to aqueous pools), even transient accumulation of small volumes of formamide in different microenvironments on early Earth such as those that could occur in rock pores or on mineral surfaces following radiolytic synthesis and dehydration could have produced segregated organic formamide microenvironments that housed chemical reactions at the microscale [42].

2.2.2. Deep Eutectic Solvents

Deep eutectic solvents (DESs) have also been recently considered as alternative nonaqueous liquids in prebiotic chemistry and could have formed readily, for example, within different ice–water systems on early Earth [48]. A eutectic solvent is a liquid made from a specific mixture of two or more substances that taken individually exist as solids, but as a mixture forms a liquid that has a single melting point lower than either of its individual components. The eutectic is the specific ratio of component compounds that exhibits the lowest melting point. A deep eutectic solvent is a mixture of solids whose melting point becomes so depressed that it exists as a liquid at room temperature. For example, a 1:2 ratio of choline chloride (melting point = 302 °C) to urea (melting point = 133 °C) has a melting point of 12 °C, and thus is a liquid at room temperature [49]. The mechanism of melting point depression is thought to involve hydrogen bonds, and so the majority of known DES mixtures involve hydrogen bond donors and acceptors [50]. DESs have the characteristics of high viscosity, low volatility, and are typically polar enough to dissolve high concentrations of ionic compounds. These non-aqueous solvents have been shown to promote various prebiotic condensation reactions including phosphorylation [51] and peptide bond formation [51,52]. Some of the components of typical DES mixtures [53,54] such as urea [55], glycerol [56], or acetamide [57] are also organic molecules thought to be generally available in prebiotic chemical systems.

2.2.3. High Pressure Supercritical Fluids (CO₂, H₂O)

One alternative non-polar environment is supercritical fluids, found in the high pressure environments of deep ocean ridges and hydrothermal systems that likely also existed on early Earth [7]. Whilst supercritical CO_2 has traditionally been thought of as rather non-polar by some in the scientific community, this, in fact, is incorrect; the polarity of supercritical CO_2 can be tuned, and CO_2 in this phase can also act either as a Lewis acid or a Lewis base depending on the specific conditions [58]. Conversely, water when supercritical becomes as non-polar as 1-dodecanol [59].

2.2.4. Tars

Non-polar environments can also be found in tars, which can be made as a product of polymerization reactions containing prebiotically available organics, and result in thick, sticky substances that exhibit extremely slow diffusion times that, even in the presence of rainfall or aqueous solutions, is practically impossible to dilute [60,61]. From a microscopic perspective, it is a reaction environment that is 'hard to leave', but offers high concentrations and extremely complex reaction environments. Some molecules, however, are able to exit through the surface by sublimation or slowly leach out into surrounding fluids.

2.2.5. Inside Lipid Bilayers and Related Interfacial Assemblies

The polar nature of water can induce amphiphilic molecules to self-assemble into a variety of phases including micelles, cubic phases, lamellar phases, and liposome or lipid bilayer vesicles. Vesicles and micelles, in particular, have been proposed to be primitive compartments that were precursors to modern cells (i.e., protocells) that could have assembled on early Earth, and the presence of such amphiphilic molecular assemblies on early Earth means that an aqueous environment can host non-polar compounds and thus non-aqueous chemistries. For a review of how these environments could affect different reactions, please see [62].

Amphiphilic molecules can also reduce surface energies [63] by adsorbing onto surfaces such as that of mineral particles [64] or exist at liquid–gas interfaces [35]. The importance of creating such layers is apparent in chemistry. Lipid monolayers have been shown to nucleate mineral growth [65], and surfaces can assist in creating desirable lipid membrane structures [66,67]. The intermembrane spaces could also be a potential site for a range of reactions, including RNA polymerization [68], where confinement to two dimensions and a non-aqueous environment is beneficial.

2.2.6. Condensed Droplet Microenvironments

Non-amphiphilic molecules can also self-aggregate via non-covalent interactions into condensed phases. These condensed phases could form due to a process known as liquid–liquid phase separation, a common phenomenon in cells that forms membraneless organelles [13]. Such phase separation could have also occurred on early Earth and would have yielded membraneless droplets that can form associatively such as coacervation between nucleic acids and cationic peptides [12,69–71], or dissociatively such as aqueous two-phase systems [72–74] or polyester microdroplets [75–77]. These droplets can host and thus concentrate molecules via similar forces as the forces that lead to condensation [78,79]. The interior of such droplet microenvironments can also vary from apolar (mainly) polymer-based environments such as in polyester microdroplets [75] to polar aqueous (but polymerrich) environments such as in coacervates [70].

2.3. *Minerals/Rocks*

2.3.1. Solid Mineral Surfaces

Solid minerals are found all over the Earth's crust in rock or suspended colloidal forms, and would have been present in abundance very early on in Earth's history. They are capable of increasing the local concentration of molecules via adsorption due to electrostatics or by reducing interfacial energies [80–83]. Mineral surfaces can also preorganize molecules while precluding water to increase reaction rates [84]. As a physical environment, minerals can contain large surface area to volume ratios, with much of the area being internal 2D interlayers such as in clays, or narrow networks of rock pores that are shielded from light as well as turbulent flow. For an overview of the importance of minerals for prebiotic chemistry, we refer readers to [85–90].

Minerals are also an important source of elements essential to prebiotic chemistry such as phosphorous [91,92]. Consequently, minerals create local microenvironments that can not only enrich molecules by adsorption, but also leach out materials to their surroundings. In particular, we note that most chemical reactions involving minerals will occur at the mineral surface, i.e., a mineral-air or mineral-water interface such as within mineral pores or cracks [87,93,94]. However, there are some mineral-based chemical processes that could occur exclusively in the solid phase, such as metamorphic changes in rocks at high temperature and pressure, which could affect the availability of certain minerals.

2.3.2. Mantle

Earth's solid mantle, which would also have formed very early on in Earth's history, is a source of minerals and gases that can partake in other chemistries once ejected/erupted onto the Earth's surface [95], and undergoes solid-state convection, a key to plate tectonics [96]. The oxidation state of the mantle is possibly driven by the disproportionation of Fe^{2+} [97], with metallic iron sinking and Fe^{3+} persisting in the mantle, rendering it oxidizing [98]. It is the high viscosity and physical inaccessibility of the mantle that enables it to be transiently out of equilibrium with the ocean, atmosphere, and crust. As a result, the oxidation state, not to mention the temperatures and pressures, can differ vastly from other regions of Earth, and enable novel (inorganic) chemistries within the mantle [99,100].

3. Physicochemical Properties

In this section, we broadly introduce the relevant physicochemical characteristics that have wide variability amongst the reaction environments introduced above. The aggregated data showing the values of each physicochemical characteristic serve to guide the design of future prebiotic chemistry studies as a way for researchers to better understand the relevance of each reaction environment to different chemical processes.

3.1. Ionic Strength

Ionic strength is, simplistically, the total concentration of charge (both positive and negative) contributed by all dissolved ions in a given solution [101]. Ionic strength contri-

butions are proportional to the square of the charge on the ion, and are thus greater for divalent ions compared to monovalent ions.

Ionic strength affects the solubility of electrolytes, inter- and intramolecular supramolecular interactions, the dissociation constant of acids (which can result in more dissolved protons in solution and lower pH [102]), and the strength of electrostatic interactions [103]. It can also impact the osmotic pressure of semipermeable systems. It may be significant that no living cell today has an intracellular concentration of 0.6 M NaCl, the sodium chloride concentration of the ocean. Instead, most cells use active transport to maintain the internal concentration of NaCl at 0.015 M, while KCl is maintained at approximately 0.15 M within the cell [104].

High ionic strengths could result in the dissociation of molecular complexes bound through charge-charge interactions such as peptide-nucleotide complexes that form primitive phase separated coacervates upon binding [69]. Salt can also inhibit the self-assembly of phospholipids into vesicles.

Here, we report the range of ionic strengths found in each of the prebiotic reaction environments introduced above (Table 1).

Environment	Ionic Strength Range (M)	References	
Aqueous solution	0.1–0.8 (oceans) 0.002–6 (lakes) 0.1–17 (lagoons) 0.1–7 (seas) 0.7–6 (hydrothermal brines)	[105]	
Sea spray	Up to 6 (marine aerosol)	[106]	
Gels	Variable, depending on components. Salt can dramatically alter gel properties.	[107,108]	
Deep eutectic solvents	Ranges from 0 to >1, but ionic strength may not be the relevant principle.	[109,110]	
Pure formamide	0.024 (commercially available pure formamide contains a significant amount of ionic impurities)	[111]	
Lipid bilayer vesicle lumens (interior)	0–0.6, depends on the solution in which the amphiphilic molecules self-assemble.	[112,113]	
Condensed droplet microenvironments	Up to 15 (within coacervate droplets)	[79]	
Solid mineral surfaces	No ionic strength for solid mineral surfaces, surface charge density may be the more relevant parameter.		

Table 1. The ionic strength of different reaction environments.

3.2. Surface Effects

When considering microenvironments, surface effects must be taken into account because of the large surface area to volume ratios of such environments compared to the bulk. Whether that interface is solid-gas, liquid-gas, liquid-solid, or liquid-liquid, the interface could be a non-negligible site that concentrates materials and increases chemical reactivity [63]. This concentration mechanism could be relevant to prebiotic chemical reactions where the reactants are highly diluted in a mixed reactant pool and would otherwise not react to any appreciable degree [114]. The reactions that occur at the airwater interface of an aerosol or droplet could therefore be more important than reactions in the bulk of the aerosol/droplet [33,115] (Figure 2).



Figure 2. In a droplet system (blue) containing pinonic acid (PA), sodium dodecyl sulfate (SDS), and hydroxyl radicals (OH) in both aqueous and gas phases (green), various reactions can occur in the droplet (volume-dominated process), the gas phase (volume-dominated process), or the gas-droplet interface (surface-dominated process). For example, SDS participates only in reactions with OH (oxidation) at surface-dominated processes due to its high surface activity (as an amphiphile). However, PA can react with OH (oxidation) both at the air-droplet interface (surface-dominated process) as well as inside the bulk droplet (volume-dominated process) due to its lower surface activity than SDS. OH can also participate in reactions in the gas phase, the liquid phase, or at the interface. Reprinted with permission from Huang, Y. et al. "Probing the OH Oxidation of Pinonic Acid at the Air-Water Interface Using Field-Induced Droplet Ionization Mass Spectrometry (FIDI-MS)". *J. Phys. Chem A.* **122**(31), 6445–6456 (2018). [115] Copyright 2018 American Chemical Society.

There are several mechanisms by which materials can accumulate at liquid interfaces. A hydrodynamic mechanism is the 'coffee ring effect'. For example, gas bubbles within heated rock pores have been shown to concentrate catalytic nucleic acids at the bubble interface and increase catalytic activity [116]. This effect can also be driven by surface tensions. Because any surface or interface has a non-zero interfacial tension at the boundary (e.g., the air-water interface being ~72.8 mN/m at room temperature), materials have a propensity to adsorb to the boundary layer, which results in an overall lower free energy. Take, for example, amphiphilic molecules, which form monolayers at the liquid-liquid interfaces that can decrease the interfacial energy by an order of magnitude [63]. Finally, surfaces can adsorb molecules directly. Those studying chemical reactions in/around rock pores need to consider the effects of the mineral surface (e.g., roughness, chemical properties) on each species participating in the reaction [93,117].

Surface features and effects are specific to each system and are not explicitly presented here. Large surface areas are also implicit in microenvironments. For more information, we refer the interested reader to the interfacial catalysis literature [118–120].

3.3. Viscosity

Chemical reactions in solution are either diffusion-limited (where reactants will react instantaneously upon contact with each other, and the reaction is thus controlled by the speed at which the reactants diffuse toward each other in solution) or reaction-limited (e.g., due to some energetic barrier) [121].

For diffusion-limited reactions, the viscosity of materials will control the speed of the reaction with higher viscosities typically slowing down reaction rates. In some cases, high viscosity may aid reactions by limiting how far molecules can diffuse from each other. Highly viscous media have been shown to support the replication and catalysis of primitive nucleic acids [48,122].

Here, we report the typical viscosities found in each of the prebiotic environments introduced above (Table 2). It should be noted that the rheology of materials (how materials deform and flow) depends on the applied stresses and strains, the temperature, and the length scales considered. Gels, for instance, can appear solid at larger length scales but still support flow inside their pores. Some materials exhibit viscous propreties at long-time scales, and elastic behavior at short-time scales.

Environment	Typical Viscosity (mPas)	References
Aqueous solution	0.89–1.00 (freshwater at room temperature) Up to 1.3 (seawater at room temperature, depending on salinity)	[123–125]
Sea spray	Ranges from 1 (sea water) to 10–10,000 during evaporation or in presence of organics	[126]
Gels	Ranges from 1 up to 2×10^6 (colloidal silica gel)	[127]
Ice	10^{15}	[128]
Deep eutectic solvents	Variable; >100 and as high as 1700 possible	[129,130]
Formamide	3.23	[131]
High pressure supercritical fluids	0.02–0.16 (CO ₂ , depending on pressure) 2.98 (water)	[132–134]
Tars	10–over 10 ¹⁰	[135]
Inside lipid bilayers	2D diffusion ~100–1000 1–1500 (heterogeneous)	[136]
Condensed droplet microenvironments	100 (coacervate)	[137]
Solid mineral surfaces	${<}1.0 imes10^{28}$ (crust)	[138]
Mantle	$2.8 imes 10^{25}$	[138]

Table 2. The viscosity of different reaction environments.

3.4. Specific Heat Capacity

Specific heat capacity is the amount of energy needed to increase the temperature of one kg of a material by one degree K. In other words, it can be used as a measure of the energy that it takes for a volume of a material to heat or cool down to a given temperature or the insulation or conduction ability of the material [139], and depends on a material's temperature and phase.

The specific heat capacity is important where temperature stability (or fluctuations) is critical. Given that the heat capacity of water (a good insulator) [140] and solids such as rocks (which could be good heat conductors) [141] are quite different, their close proximity in the form of water-rock interfaces (such as in hydrothermal vent environments or hot springs [142]) leads to significant heat transfer and could potentially affect processes such as self-assembly [87], geoelectrochemistry [143], transport in thermal gradients [30], evaporation, or even mineral composition [144].

Here, we report on the range of specific heat found in each of the prebiotic environments introduced above (Table 3).

Environment	Specific Heat (kJ/Kg K)	References
Aqueous solution	4.18 (freshwater) 3.6–4.18 (saltwater), at room temperature.	[124,125,145,146]
Sea spray	Aerosols readily evaporate; specific heat is	not very relevant.
Gels	0.8–1.10 (silica gel) Specific heat for hydrogels depends on water level and temperature, for example, up to 30.	[147,148]
Ice	0.4873–0.3496 (from 0 to -80 °C, respectively)	[149]
Deep eutectic solvent	1.5–1.8 (example of salt eutectic)	[150]
Formamide	2.39	[151]
High pressure supercritical fluids	3–30 (CO ₂ , depending on pressure) 27–690 (water, depending on pressure)	[152,153]
Tars	1.25–2	[154]
Inside bilayers	0.3–0.9; higher near melting temperature	[155,156]
Condensed droplet microenvironments	1.483	[157]
Solid mineral surfaces	0.180 (bromyrite) to 1.510 (epsomite); however, most are between 0.3 and 0.9	[158]
Mantle	1.250	[159]

Table 3. The specific heat of different reaction environments.

3.5. pH

Because pH is defined as a solution property, only liquids can exhibit a pH. pH is usually defined in terms of the autoionization of water, but the concept of pH can also be extended to neat nonaqueous solvents as long as they have some ability to donate a proton [160]. The solution pH affects chemical properties such as the protonation state of molecules in the solution and hence their potential to participate in chemical reactions or assembly into supramolecular structures. For example, pH fluctuations could give rise to the cyclical assembly and disassembly of coacervate droplets due to changes in the charge states of the constituent polymers [161], while pH changes also modulate RNA base-pairing, resulting in the ability to affect strand separation [162] and vesicle self-assembly [163]. While the pH of a solution is generally uniform, there are some cases where the pH within an environment exhibits changes and is not uniform such as in certain terrestrial lakes [164] or water mixing zones [165].

Here, we report on the range of pH found in each of the prebiotic environments introduced above (Table 4).

Table 4. The	pH range of	different reaction	environments.
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Environment	pH Range	References		
	6.3–7.2 (4.0 Ga ocean)			
	6.5–7.7 (2.5 Ga ocean)			
	8.2 (modern ocean)			
	6–8 (freshwater)			
Aqueous solution	Pure water is 7.0	[4,166–168]		
-	Hot spring environments have more			
	variability, and can range from very			
	acidic (less than pH 3) to somewhat			
	alkaline (as high as pH 10).			

Environment	pH Range	References	
Sea spray	Around 8.0	[169]	
Gels	Variable, depending on components.	[75,170–173]	
Deep eutectic solvents	1.2–13.5 (eutectic at room temperature; pH varies greatly between eutectics, and also changes with temperature, down to pH 0)	[174,175]	
High pressure supercritical fluids	2.80–2.95 (of water around scCO ₂)	[176]	
Inside lipid bilayers	pH can be of a variety of ranges such as low as pH 2 or lower [177] or as high as pH 12 [178].		
Condensed droplet microenvironments	Highly dependent on the components, and especially their charge states at different pH (i.e., pKa).		
Solid mineral surfaces	Aqueous solutions containing solid mineral surfaces are mostly acidic. However, some have been found that were alkaline (pH 8.7–9.6).	[179]	
Mantle	Mantle-derived igneous rocks can be alkaline, while mantle-derived minerals on the seafloor (around hydrothermal systems) can be around pH 9–11	[180,181]	

3.6. Density

The density of reaction environments is important to consider, because differences in density could lead to the physical separation of different components, a process that occurs during ocean stratification or hydrodynamic sorting.

While solids are generally more dense than liquids, which are more dense than gases, there are cases where this is inverted such as tungsten hexafluoride gas [182] being at least 10 times denser than graphene aerogel solid [183] or solid pumice being able to float on water. The density of a material will increase upon increasing pressure (decreasing the volume due to pressure-driven compression (Section 3.10)), while increasing temperatures will usually, but not always [184], cause a density decrease. Some materials are non-uniform (e.g., rocks or minerals [185]), leading to different microenvironments even within the same material.

Here, we report on the density of each of the prebiotic environments introduced above (Table 5); however, as there are different environments on early Earth with variable temperatures (such as hot springs [6]) and pressured (such as near hydrothermal vent systems in the deep ocean [186]), the densities reported here may change accordingly.

Table 5. The density of the different reaction environments.

Environment	Density (g/mL)	References
Aqueous solution	0.9999749 (freshwater at 4 °C); 0.9970470 (freshwater at 25 °C) 1.025 (seawater, average; can be up to 1.09 depending on salinity)	[124,125,187]
Sea spray	1.12–2.16 (at room temperature)	[188,189]
Gels	Lower bound is that of the solvent for dilute gels.	
Ice 0.84–0.91 (sea ice)		[190]
Deep eutectic solvent	0.8–1.8 (example of a eutectic between 5 and 100 °C)	[174,191]

Environment	Density (g/mL)	References
Formamide	1.129 (at 25 °C)	[131]
High pressure supercritical fluids	0.1–1 (CO ₂ , depending on temperature and pressure) ~0.1–0.326 (water, depending on temperature and pressure)	[134,192,193]
Tars	1.1–1.23	[194,195]
Lipid bilayers	~0.9 for the lipid bilayer itself (e.g., decanoic acid density is 0.893 g/cm ³) In the aqueous lumen, values as per 'aqueous solution'.	[196]
Condensed droplet microenvironments	1.18–1.92	[197]
Solid mineral surfaces	1.2 (kerogen) to 10.969 (uraninite); however, most are typically between 2 and 7	[158,198]
Mantle	3.4 (mantle surface, and gets larger deeper)	[198]

3.7. Dielectric Constant

A general adage in chemistry is that "like dissolves like"; polar solvents are more likely to dissolve charged solutes or solutes with high dipole moments (i.e., polar compounds) [199]. This is because the ability of a solvent to disrupt solute-solute interactions depends on the specific intermolecular forces involved.

One parameter used to estimate solvent polarity is the zero-frequency component of the dielectric constant (ε). While other measures such as hydrogen bonding capacity, dipole moment, and acidity/basicity are also important, the dielectric constant remains a good rule of thumb for estimating the polarity of the solvent as well as how miscible solvents are with each other [200]. Formally, ε is the relative permittivity of a material compared to vacuum and is defined as the amount of polarization that a material will experience (i.e., the magnitude of dipole moments) when an electric field is applied to it [199,201]. This means that ε is a measure of the polarizability of a solvent, with solvents having $\varepsilon \gtrsim 10-20$ defined as polar. As points of reference, apolar organic solvents have a relatively low $\varepsilon \sim 2$, while polar water has an $\varepsilon \sim 80$ [202]. Solvents with similar dielectric constants are generally miscible.

Given the wide variety of chemistries thought necessary for the origin of life [203], it could have been possible for a variety of aqueous and nonaqueous media to contribute to the potential prebiotic reaction space. Polar solvents can be divided further into protic solvents (able to hydrogen bond or donate hydrogen) and aprotic solvents, which can be determined by looking at the solvent molecule's structure.

Here, we report the ε of each of the prebiotic environments introduced above (Table 6).

Table 6. The dielectric constant	t (ε) of	different	reaction	environments
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Environment	ε (unitless)	References
Aqueous solution	~70–80 (decreases with increasing temperature and salinity; seawater may be slightly lower than freshwater)	[204,205]
Sea spray	2.5–50	[206]
Gels	1.008–1.9 (silica gel, depending on density)	[207]

Environment	ε (unitless)	References
Ice	30–130 (ice)	[208]
Deep eutectic solvent	22.8 (one example)	[109]
Formamide	105–113 (room temperature)	[209,210]
High pressure supercritical fluids	1.07–1.46 (CO ₂ , depending on temperature and pressure)	[211]
Tars	Up to 8 (coal tar)	[195,212]
Inside bilayers	2–3, can be higher for membranes that are more permeable than phospholipids	[213,214]
Condensed droplet microenvironments	40–50	[215]
Solid mineral surfaces	4.9–7.5	[216]
Mantle	~38 (water in the upper mantle at 300 km and 1000 K)	[217]

Table 6. Cont.

3.8. Boiling, Melting/Freezing Temperatures

At higher pressures such as in the deep ocean [7], different phase transitions can occur such as the direct sublimation of ice to water vapor upon increasing temperature [218]. Additionally, hysteresis, such as in rock pores, has also been observed (i.e., the freezing temperature is not identical to the melting temperature [219]).

Knowing the phase transition temperatures of materials is important for several reasons. Phase transitions accessible to the temperatures and pressures on early Earth impact the abundance of solvents. Additionally, materials in different phases have very different properties. Carbon dioxide gas and supercritical liquid carbon dioxide will have different affinities for various prebiotically plausible chemicals [203,220,221], resulting in differences in the reactivities or plausible chemistries residing within such environments. Furthermore, it has been shown that freeze-thaw cycles in water could have contributed to primitive genetic biopolymer (i.e., RNA) replication and assembly [222,223].

Here, we report on the boiling (liquid to gas transition) and melting (solid to liquid transition) temperatures of each of the prebiotic environments introduced above at atmospheric pressure as a point of reference (Table 7). However, as there were different environments on early Earth with variable pressure such as near hydrothermal vent systems in the deep ocean [186], the temperatures reported here will change accordingly (and at pressures below the triple point, there may only be one phase-transition temperature, i.e., sublimation, physically possible). Additionally, for some systems such as condensed droplet microenvironments, "melting" may refer to the transition from the condensed phase to the uniform phase, as increasing temperatures will inhibit the non-covalent bonds required for the structure to form, and depends on the composition of the system [69,224,225].

Table 7. The boiling and melting/freezing temperatures of different reaction environments.

Environment	Boiling Temperature	Melting/Freezing Temperature	References
Aqueous solution	Freshwater (100 °C); As high as 102 °C (seawater, depending on salinity)	Freshwater (0 °C); As low as –2 °C (seawater, depending on salinity)	[124,125,226–228]
Sea spray	70–100 °C	Close to 0 °C	[229,230]
Gels	2230 °C (silica gel)	1710 $^{\circ}$ C (silica gel)	[231]

Environment	Boiling Temperature Melting/Freezing Temperature		References
Ice (eutectic)	In solid form, same as water (depending on salinity).		
Formamide	210 °C	2–3 °C	[232,233]
High pressure supercritical fluids		See footnote *	
Tars	190–400 °C		[234]
Inside lipid bilayers		See footnote ^	
Solid mineral surfaces	N/A	700–900 °C	[235]
Mantle	N/A	~3600 °C near the core–mantle boundary	[236]

Table 7. Cont.

* Typical "boiling" and "melting/freezing" transitions may not be applicable. Rather, the supercritical fluid to liquid, solid, and/or gas transition temperatures will depend on the pressure and is unique to each system based on the phase diagram. For example, scCO₂ will transition to the liquid state below 304 K at 100 bar, but will transition to the solid state below 304 K at 10,000 bar [220]. Supercritical water will transition to the liquid state below 647 K at any pressure above 22.1 MPa; supercritical water cannot directly transition to the solid form under any circumstances [237]. Neither scCO₂ nor supercritical water can transition to the gas phase based on temperature changes and can only transition to the gas phase upon decreasing pressure. ^ The lipid bilayer itself may not boil (as boiling requires the bilayer to vaporize, effectively resulting in the loss of the bilayer structure). However, the boiling point of the lipids that compose the bilayer vary depending on lipid composition. Typically, the boiling point of 205.8 °C, while stearic acid (C18 saturated) has a boiling point of 376.1 °C [238]. The "melting" of a bilayer refers to the solid (gel) to liquid transition, and not the melting of the lipid components themselves. This also depends on the lipid composition; longer chain lipids typically have a higher phase transition temperature [239,240].

3.9. Vapor Pressure

Vapor pressure is related to the volatility of a material (i.e., the amount of gas that is released from a material at any given point) with the boiling point being defined as when the vapor pressure of the liquid material is equivalent to the ambient pressure (Section 3.8). Extreme cases include the highly volatile ammonia [241] and non-volatile mineral oil [242]. Higher temperatures will result in higher vapor pressure as per the Antoine relation [243]. Vapor pressure is also applicable to solids that sublimate (e.g., dry ice) [244]. A related concept is Henry's law for mixtures of gases, which relates the amount of a dissolved gas to the partial pressure of that gas.

The volatility of a prebiotic material impacts whether that reaction environment is stable at a given temperature, or whether it will spontaneously (and quickly) change form into a gas, even below the boiling point. For example, it has been reported that the vapor pressure of fatty acids [245] and fatty acid esters [246] decreases with increasing chainlength. This suggests that fatty acids on early Earth, which could undergo liquid-phase reactions at high temperatures such as in hot spring environments [6], may have been more biased toward longer-chain fatty acids, as shorter chain fatty acids would likely have been volatilized into the gas phase.

Here, we report on the vapor pressure of each of the prebiotic environments introduced above (Table 8).

Environment	Vapor Pressure (kPa)	References
Aqueous solution	2.3–4.2 (freshwater, room temperature) 2.1–3.9 (seawater, room temperature, depending on salinity)	[124,125]
Gels	~0.13–2.3, depending on the gel formulation and conditions.	[247]

Table 8. The vapor pressure of different reaction environments.

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Environment	Vapor Pressure (kPa)	References
Ice	6.1 (ice at 0 °C), but decreases with decreasing temperature (for example, 0.1 at -20 °C and 0.0014 at -100 °C).	[248]
Deep eutectic solvent	1.48 (CaCl ₂ eutectic in water at 20 °C). However, vapor pressure of other eutectics may vary depending on composition and temperature.	[249,250]
Formamide	Formamide 0.008	
Inside bilayers	Vapor pressure will be related to the vapor pressure of the bilayer components; vapor pressure typically decreases with increasing chain length (at constant temperature).	[251,252]
Condensed droplet microenvironments	Very low to negligible vapor pressure (ionic liquids)	[253]
Solid mineral surfaces	Around 0.05–0.25 (melted minerals >1900 K) Vapor pressure of solid mineral surfaces is negligible	[254]

3.10. Compressibility and Stiffness

For solids, one measure of deformability is the Young's Modulus (*E*) (Figure 3a). Materials with lower *E* are more compressible (less stiff), and vice versa. The *E* of coal is about 10 times less than limestone [255,256]. The Young's modulus is only applicable to solids, as fluids (such as liquids and gases) require zero force to change in size lengthwise.

Bulk modulus (*K*) is defined as the amount of pressure (equally from all sides) required to effect a resulting volume change on a material (Figure 3b) and is defined for both fluids and solids. A typical gas has a very low *K* of <0.1 GPa [255] whereas quartz or clay has a *K* of 20–40 GPa [255].

While not immediately obvious, the compressibility and stiffness of materials become relevant on early Earth either in high-pressure environments, or when environments encounter high pressures resulting from a large external force. Examples include pressure from water deep in the ocean [186] or the late heavy bombardment during impacts [257]. The temporary deformation or compression of surface minerals with low *E*, for example, could have affected primitive mineral-driven processes such as mechanochemical sugar [258] or peptide [94,258] synthesis, possibly within mica sheets [259].

Here, we report the *K* and *E* of each of the prebiotic environments introduced above (Table 9).

Table 9.	The	Young's	(E)) and	bulk	(K)	modulus	of	differe	ent	reaction	enviror	ments
			· · · ·			× /							

Environment	E (GPa) *	K (GPa)	References
Aqueous solution	-	2.1	[260]
Gels	0.05–10 (of a silica aerogel, depending on gel density)	4–20 (of an alkaline-calcium silica hydrogel, depending on pressure)	[261,262]
Ice	8.6–12 (depends on the plane)	8.5–11.5 (depends on temperature)	[263,264]
High pressure supercritical fluids	-	1 (water at room temperature and pressure) 0.1–0.7 (CO ₂ , depending on temperature and pressure)	[192,265,266]
Inside bilayers	0.02–0.03	0.6–0.9 (depending on temperature and location)	[267–269]

Environment	E (GPa) *	K (GPa)	References			
Condensed droplet microenvironments	These values will all dependent	These values will all depend on the droplet composition; "aging" is also an issu these droplets.				
Solid mineral surfaces	6.38–288 (depending on the mineral and pressure)	40–120 (depending on mineral and pressure)	[270–272]			
Mantle	150–720 (depending on depth)	100–600 (depending on depth)	[272]			

Table 9. Cont.

The Young's modulus is not defined for liquids and gases.



Figure 3. A physical description of the direction of forces used to calculate (**a**) the Young's modulus (*E*) and (**b**) bulk modulus (*K*). Reprinted with permission from Burtch, NC, et al. "Mechanical Properties in Metal-Organic Frameworks: Emerging Opportunities and Challenges for Device Functionality and Technological Applications". *Adv. Mater.* **30**(37), 1704124 (2018). [273] Copyright 2018 Wiley.

3.11. Exposure to Radiation

Radiation is the process of energy transmission in the form of photons or massive particles and includes electromagnetic radiation (radio waves, microwaves, infrared, visible light, ultraviolet, X-rays, and gamma radiation) and particle radiation (e.g., alpha and beta particles, neutrons). Electromagnetic waves carry radiant energy as photons wherein the wavelength determines the energy, whereas particle radiation is the result of fast-moving subatomic particles whose energies can vary depending on their mass and velocities. Radiation is further classified as non-ionizing versus ionizing depending on the energy of the photon or particle. The energy of ionizing radiation is broadly defined to be between 10 and 33 eV, which is typically enough to ionize molecules and break chemical bonds (the energy of a C–C bond, for example, is about 3.6 eV). The region of electromagnetic radiation including higher energy ultraviolet, X-rays, and gamma radiation as well as typical particle radiation (alpha radiation, beta radiation and neutron radiation) are all considered types of ionizing radiation.

On early Earth, UV radiation would have been a strong driver of chemical synthesis and evolution. While the presence of ozone on modern Earth can absorb all ionizing and 98% of non-ionizing UV light, the surface of early Earth was exposed to much higher fluences of UV light (in particular, wavelengths longer than ~200 nm) prior to the build-up of atmospheric oxygen and thus ozone. In addition, while the young Sun was about 25% less luminous than today [274], its output in the UV region was likely larger. UV radiation has been demonstrated to play a key role in the synthesis of prebiotic molecules [275]. For example, the Sutherland group reported a UV radiation-driven photosynthesis of simple sugars from HCN [276], while UV radiation has also been shown to produce amino acids in the atmosphere [277]. However, UV radiation is also a double-edged sword, and has been shown to induce the degradation or structural/configurational alteration of organic molecules including DNA [278], proteins, and lipids [279].

Radiolysis from ionizing radiation is also employed in prebiotic chemistry as a synthetic mechanism [57]. Ionizing radiation could have been present through multiple sources such as cosmic rays and radioactive minerals. Cosmic rays consist of high-energy protons and atomic nuclei originating from the Sun or outside of the Solar System [280]; the energy spectrum of primary cosmic rays is between 10^{16} eV (2.31 \times 10¹⁷ kcal/mol or 3.89 \times 10¹⁷ kT, at 298 K) and 10^{18} eV (2.31 \times 10¹⁹ kcal/mol or 3.89 \times 10¹⁹ kT, at 298 K) [281]. Cosmic rays impact Earth's upper atmosphere to produce showers of secondary photons and particles. A higher output of solar energetic particles from the young Sun via more frequent solar flares and coronal mass ejections has been proposed to have exposed the atmosphere of the early Earth to significantly higher fluxes of radiation in comparison to today [282]. Ancient radioactive mineral deposits containing, for example, monazite and uraninite [283], could also have provided locally high fluxes of alpha, beta, or gamma radiation on their surfaces [284]. Under the primordial conditions of the Hadean eon when the ²³⁵U isotope was much more abundant, so-called natural nuclear reactors that can promote self-sustaining nuclear fission chain reactions may have been relatively commonplace and produced highly radioactive local environments [285]. Over a dozen individual fission zones are known from the Oklo locality in Gabon [283]. A typical fission zone comparable in size to those found at Oklo are thought to produce ~10 kilowatts of power output (radiolysis and heat) wherein ~13% of this power is composed of γ or β rays that can penetrate substantial distances beyond their host minerals [283,284].

The availability of radiation in prebiotic reaction environments is determined by how far the radiation is able to penetrate a given material and the characteristics of the radiation source. Gamma radiation, arising from the radioactive decay of atomic nuclei, is characterized by short-wavelength electromagnetic waves ($\sim 10^{-11}$ m) with the highest photon energies above 100 keV. Gamma radiation is capable of penetrating materials to significant depths, requiring thick layers of high-density materials to block it. In comparison, alpha radiation with a fast-moving helium-4 nucleus is halted by a sheet of paper, and beta radiation consisting of high-velocity electrons can be stopped by an aluminum plate. The spectrum of the young Sun and attenuation of UV light by gases and aqueous media is covered thoroughly by Ranjan and Sasselov [275].

4. Conclusions

Here, we introduced some important physicochemical properties of various prebiotic reaction environments, with some brief examples of relevant prebiotic processes that could have been modulated by those physicochemical properties. Correlating physicochemical properties with reaction environments, while considering such properties during the design of prebiotically plausible reactions is highly relevant to the origins of life field. However, values of physicochemical characteristics in some environments still remain to be elucidated. This work serves as a request for the community to contribute to "filling in the blanks" in future works, so that researchers in the field can have a more holistic understanding of the prebiotic geochemical reaction environment. We also acknowledge the fact that experiments that fill in these blanks could be rather tedious and may not lead to high-profile publications; however, such studies are essential to pursue.

From this overview, it is evident that an extremely wide range of physicochemical conditions can be accessed through the early Earth environment, which further supports the widely accepted notion that a large repertoire of chemical reactions were taking place on early Earth. Likewise, the prebiotic chemical repertoire is also highly diverse, with reactions or processes that may have had a range of tolerances to a wide variety of conditions and others that did not. The fact that modern biology, especially in the form of extremophiles, is also tolerant to a wide variety of conditions could be an artifact of the chemical reactions or processes that led to the life's origins.

Generally speaking, the robustness/tolerance of the entire prebiotic chemical reaction repertoire to changes in conditions is at present not well-understood due to the large parameter space. In future, probing the "limits" of a wide range of prebiotic processes is necessary to gain a better understanding of which prebiotic environments could have plausibly hosted certain prebiotic chemical reaction networks. By aggregating this type of data, it will also be possible to conjecture which groups of prebiotic chemical processes/reactions could have been co-localized with each other as well as those which likely could not have occurred simultaneously under the same conditions in the same location.

The environments, properties, and examples of prebiotic processes provided here are but a brief and general overview of the entire prebiotic chemical milieu, and are not meant to be an exhaustive or comprehensive resource. A number of parameters for many primitive environments are not known (and as such, cannot be presented here), which behooves the field to continue characterizing the unknown physicochemical properties of all reaction environments. We further look forward to the field's continuing exploration of extraterrestrial reaction environments, which could also provide insights into the possible reaction conditions on early Earth.

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