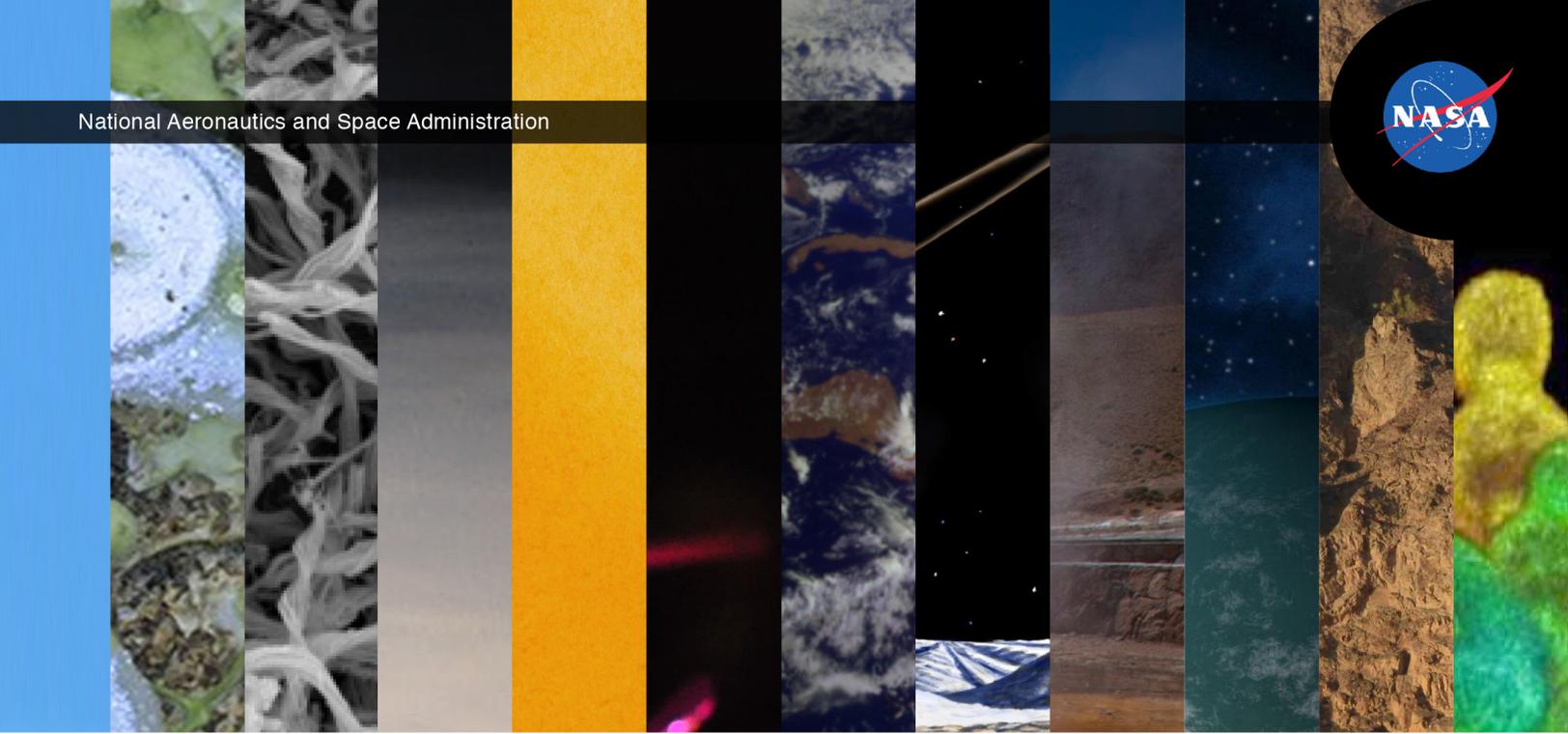
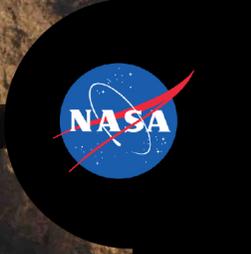


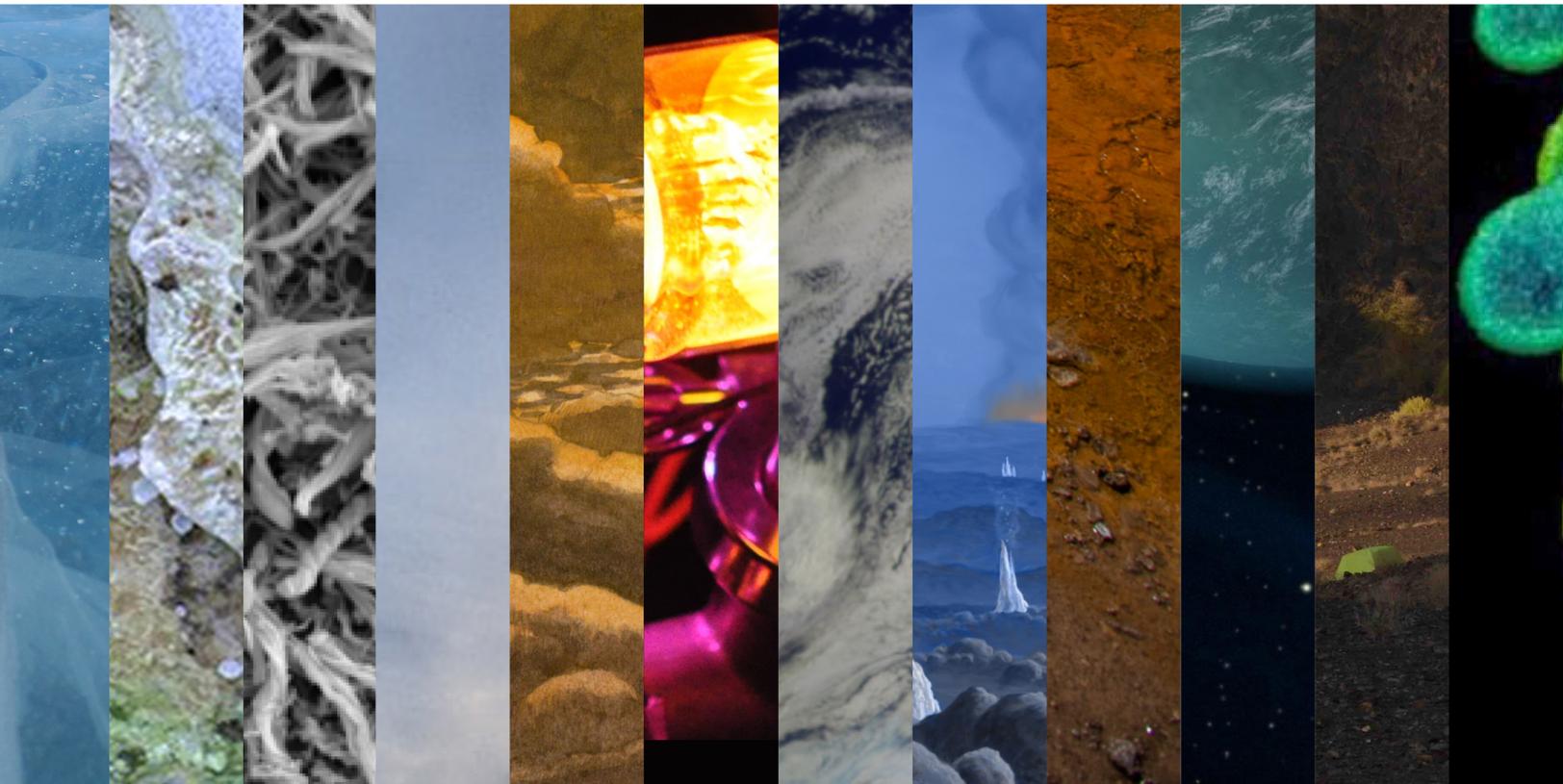
National Aeronautics and Space Administration



NASA ASTROBIOLOGY INSTITUTE

2017 Annual Science Report

Icy Worlds: Astrobiology at the Water-Rock Interface and Beyond
NASA Jet Propulsion Laboratory





Icy Worlds: Astrobiology at the Water-Rock Interface and Beyond

Lead Institution:
NASA Jet Propulsion Laboratory



Team Overview



Principal Investigator:
Isik Kanik

Astrobiology at water-rock interfaces found on icy bodies (e.g., Europa, Enceladus and Ganymede) in our Solar System (and beyond) is the unifying theme for the proposed research. In this interdisciplinary research, our Team (The Icy Worlds: Astrobiology at the Water-Rock Interface and Beyond) conducts a highly synergistic combination of experimental, theoretical, and field-based lines of inquiry focused on answering a single compelling question in astrobiology: How can geochemical disequilibria drive the emergence of metabolism and ultimately generate observable signatures on icy worlds? Our team's primary goal is to answer one of the most fundamental questions in all of astrobiology: What geological and hydrologic factors drive chemical disequilibria at water-rock interfaces on Earth and other worlds? Our research encompasses four investigations (INV's):

- What geological and hydrologic factors drive chemical disequilibria at water-rock interfaces on Earth and other worlds?
- Do geoelectrochemical gradients in hydrothermal chimney systems drive prebiotic redox chemistry towards an emergence of metabolism?
- How, where, and for how long might disequilibria exist in icy worlds, and what does that imply in terms of habitability?
- What can observable surface chemical signatures tell us about the habitability of subsurface oceans?

Team Website: <https://icyworlds.jpl.nasa.gov>

2017 Executive Summary

INV 1 team has continued to work to establish a relationship between the geochemistry of moderate temperature serpentinization with that of early biochemical pathways (i.e. biochemical methanogenesis derived from a rapid and facile geochemical reduction of CO_2 to CH_4). The big surprise in INV 1 was that only formate, and not methane, was produced during moderate temperature serpentinization. This led us to consider that methane in pristine alkaline hydrothermal solution would have been the abiotic 'fuel' rather than the 'exhaust' of the supposed first cells (Russell and Nitschke, 2017). Based on this new approach, we calculated the likely concentration of nitrate required to be present in the Hadean Ocean as reported in Wong et al., 2017. We also continued our investigations on "Green Rust" which would have been the main precipitate at Hadean submarine alkaline vents (Russell, 2017). Green Rust appears to be most promising for incubating life for providing the needed disequilibria conversion engines or engines of Creation (Branscomb et al., 2017). Because our research findings, as to be expected of the alkaline vent theory, we felt confident in applying the model to Europa (Fig.1), known to have an irradiated and oxidized surface (Russell et al., 2017).

INV 2 team has continued to working on understanding amino acid synthesis, nitrogen chemistry, and organic/phosphorus retention in hydrothermal minerals on early Earth, and electrochemical characterization of simulated hydrothermal systems. Significant effort was dedicated to the synthesis of pure green rust (GR) samples, which are highly air sensitive and need to be prepared using strict anaerobic techniques (Fig. 2). The GR-mediated reduction of mixed nitrate/nitrite systems was investigated in the current Project year. Originally, analytical method sensitivity was a problem, and so subsequent efforts focused on using colorimetric methods to characterize N and Fe species. We have also dedicated significant effort to investigating the synthesis of amino acid driven by hydrothermal iron hydroxides, as a function of the redox / pH / temperature gradients within the system. This work is being prepared for publication, and we are beginning the next steps which are to investigate chirality of the amino acids produced and to link amino acid synthesis with GR-driven nitrogen chemistry. We also have continued investigating phosphorus chemistry in GR and iron hydroxide mediated systems. Future work will investigate coupling these N and P chemistries in green rust systems to organic synthesis. We also have been working on the development of new methods to characterize redox processes at prebiotic vent systems, and on how the addition of organics changes the structure and growth of hydrothermal chimneys. New types of experiments developed this year include multi-electrode arrays for characterizing hydrothermal chimney electrochemistry and new reactors for temperature gradient testing.

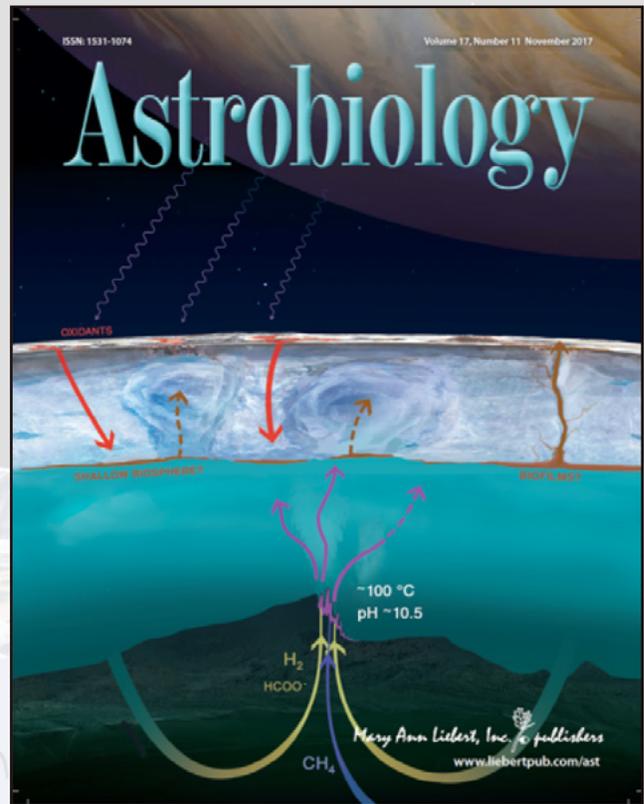


Fig. 1. Model for the emergence of life in the European ocean at an alkaline hydrothermal mound. The cells that first evolved from this hatchery metabolized hydrogen, methane, carbonate, nitrate, nitrite, ferrous and ferric iron, and phosphate. The nitrate, nitrite and ferric iron would be rapidly depleted by these metabolists. Replacement of these oxidants from the icy exterior, produced through intense radiation from Jupiter and gravitating to the base of the ice-lid, is slow. Therefore, only those cells entrained within buoyant thermal plumes in the ocean and fortuitously finding their way to oxidant zones at the base of the ice shell, could dispose of their waste electrons and survive.

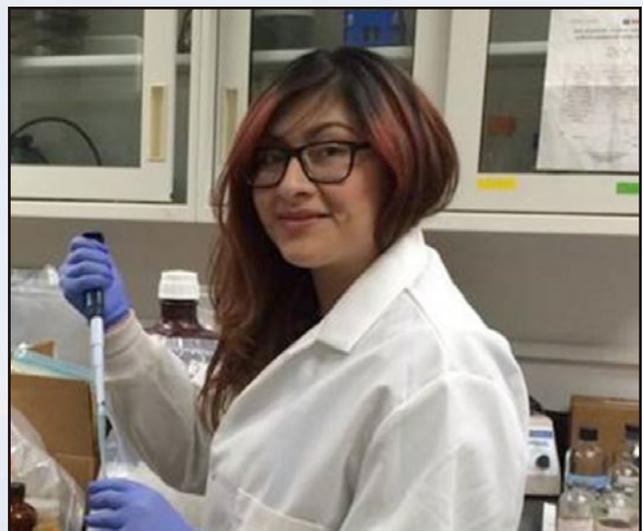


Fig. 2. Erika Flores, a NASA graduate Intern with the NAI Icy Worlds team, is working on the synthesis of pure green rust (GR), a precipitate formed from adding Fe^{2+} and Fe^{3+} to a sodium hydroxide solution, to be used for investigating phosphorus chemistry in GR and iron hydroxide mediated systems.

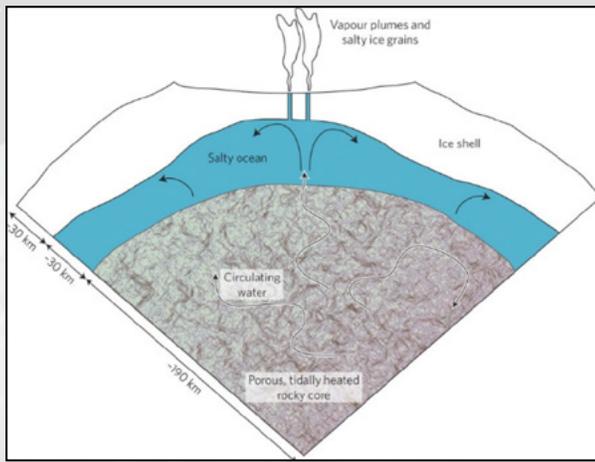


Fig. 3. A new study by the NAI Icy Worlds team predicts that tidal friction could generate more than 10 GW of heat which not only favors to maintain a liquid ocean, intense water-rock interactions and the transport of hydrothermal vent products from the core of plume source but it is also enough to power hydrothermal activity for tens of millions to billions of years inside Enceladus. Image Credit: Nimmo, F., (2017) Nature Astronomy, 1: 821.

INV 3 which looks at how, where, and for how long might disequilibria exist in icy worlds, and what that may imply in terms of habitability. In year 3, we continued to examine how geophysical investigations by future spacecraft missions such as Europa Clipper can yield new information about habitability (Vance et al. 2017a, b). We continued to develop our analytical model for ice-ocean interactions in Europa and other icy satellites by incorporating the ocean's salinity effects and variations in heat flow analogous to Earth's thermohaline circulation. We also examined the likely porosity of Europa's near surface. The results show that previous studies had overestimated the likely porosity which has major implications for the nature of heat and material transport through the surface and underlying ice and ocean. Team members at the University of Washington, Seattle, continued their innovative work examining the behavior of fluids at very high pressures found in the larger icy satellites. INV 3 team members completed two studies (Choblet et al., Nature Astronomy, 2017;

Kalousova et al., Icarus, 2018). The first study, based on a numerical model, investigated the amount of tidal dissipation that can be produced in the porous silicate core of Enceladus. The model proposes a tidally heated core, driving hydrothermal circulation, that the amount of tidal heat can be several 10s of GW, which can explain how an ocean can persist during billions of years in such a small body (Fig. 3). This research would increase Enceladus' potential as a habitable world. If the required conditions have been sustained long enough, the likelihood would increase for life to emerge within this distant icy world. The second study deals with the dynamics of the high-pressure ice layer predicted to exist in Ganymede's interior between the ocean and the silicate core. We continued to seek to connect our investigations of ocean worlds in the solar system with possible habitable ice-covered exoplanets (HICEPS) that probably exist in the current lists of exoworlds.

In Year 3, INV 4 researchers have continued to work on to answer the Key Question of "What can observable surface chemical signatures tell us about the habitability of subsurface oceans?" in order to shed light on the evolution of ocean materials expressed on the surface of airless icy bodies and exposed to relevant surface temperatures, vacuum, photolysis and radiolysis. To this end, an experimental program, designed to establish the extent to which chemical compositions of icy world surfaces are indicative of subsurface ocean chemistry, has been initiated. The initial experiments focused on freezing solutions of sodium, magnesium, sulfate and chloride – four commonly suggested major components of Europa's Ocean. In conjunction, solutions containing sodium, ammonium, chloride, and carbonate, which have been implicated in the formation of Ceres' bright spots have been investigated (Fig. 4) which provide evidence that (i) ammonium-bearing materials are kinetically-favored products under fast-freezing conditions with these brine mixtures, and (ii) the bright spots on Ceres's surface likely originate from subsurface liquids, mobilized either by a cryovolcanic process or by the Occator-forming impact, and not by direct excavation.

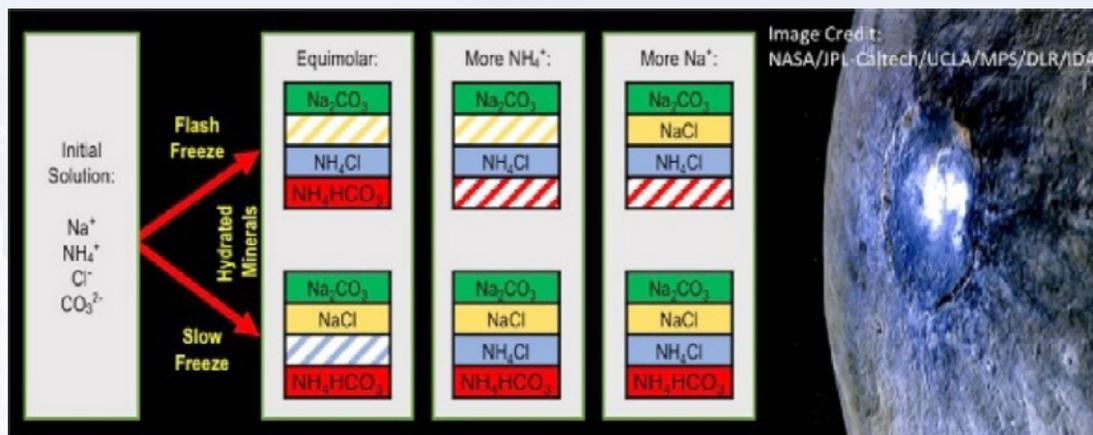


Fig. 4. Kinetic effect on the freezing of ammonium-sodium-carbonate-chloride brines and formation of minerals on Ceres' bright spots containing natrite (Na_2CO_3) and smaller amounts of NH_4Cl or NH_4HCO_3 .

Project Reports

INV 1 - Geochemical Reactor: Energy Production at Water-Rock Interfaces

In this investigation, we examine water-rock interactions to characterize the geochemical gradients that could be present at water-rock interfaces on Earth and other worlds, taking into account different ocean and crustal chemistries. The big surprise in INV 1 was our finding that only formate, and not methane, was produced during moderate temperature serpentinization. This led us to consider that methane in pristine alkaline hydrothermal solution would have been the abiotic 'fuel' (for the first methanotrophs) rather than the 'exhaust' of the supposed first cells, i.e., the methanogenic metabolism must have evolved after life first emerged (Russell and Nitschke, 2017)! This new model required the high potential electron acceptor nitrate. Thus, it behoved us to calculate the likely concentration of NO in the Hadean atmosphere, which led us to conclude that the NO concentrations in the atmosphere, and thereby the nitrate concentration in the Hadean Ocean met the assumptions of the model (Wong et al., 2017).

With the knowledge that green rust was capable of donating 8 electrons to nitrate and thereby rapidly reduce it to ammonia, we recognized ammonia's potential to aminate carboxylic acids to amino acids such as alanine (Flores et al., 2017, ISSOL), and that within such fine interlayers the alanine would polymerize to a heterochiral poly-alanine – a peptide capable of sequestering inorganic clusters. Realizing the power of green rust to act as a nanoengine, converting disequilibria to drive endergonic reactions (Fig. 5), gave us the motivation to write "Escapement mechanisms and the conversion of disequilibria: The engines of creation" (Branscomb et al., 2017). That green rust would have been the main precipitate (along with silica and minor iron sulfides) at Hadean submarine alkaline vents was argued in Russell (2017).

Because our research findings were in concert with the expectations of the alkaline vent theory – particularly for the requirement for high potential electron acceptors, we felt confident in applying the model to Europa, known to have an irradiated, and thereby oxidized surface (Russell et al., 2017).

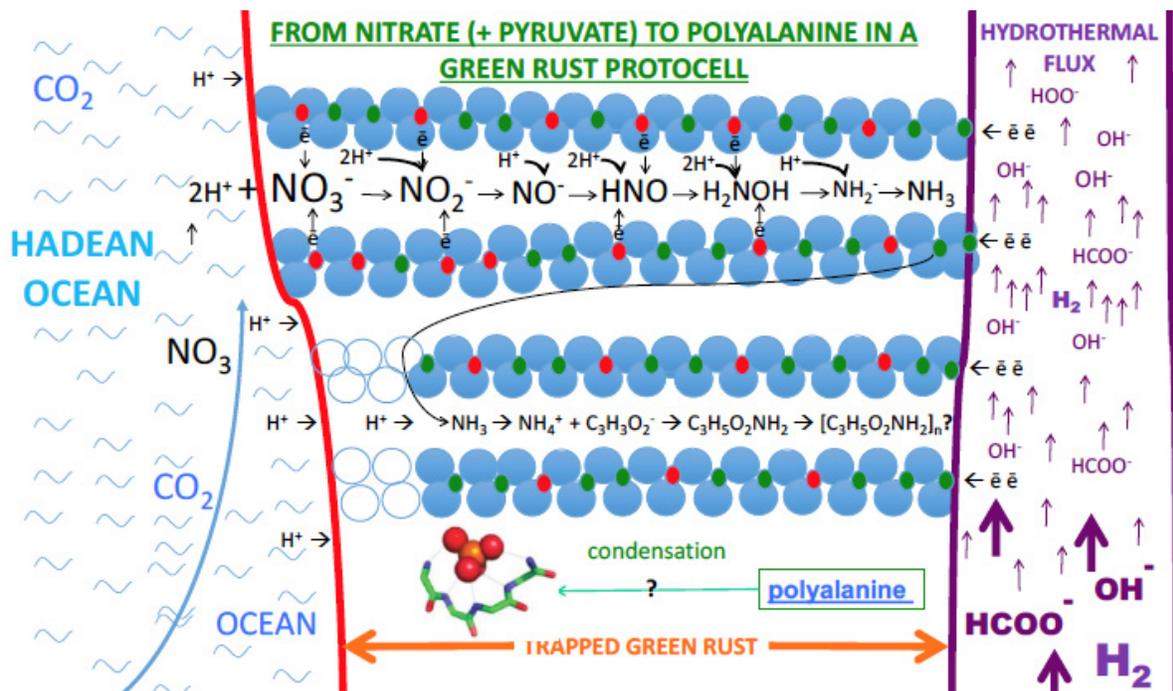


Fig. 5. A green rust ramjet reducing nitrate to ammonia and then aminating pyruvate to polyalanine (Russell and Nitschke, 2017; Branscomb et al., 2017; Flores et al., 2017)

INV 2 - From Geochemistry to Biochemistry

INV 2 focused on understanding amino acid synthesis, organic/phosphorus chemistry, green rust (GR) synthesis, and nitrogen chemistry in hydrothermal systems. Significant effort was dedicated to the synthesis of pure GR samples. These are highly air sensitive and need to be prepared using strict anaerobic techniques. During the current Project year, a number of GR species were synthesized, including GR1(F-), GR1(Cl-), GR1(Br-), GR1(CO₃²⁻), and GR2(SO₄²⁻). The GR species were characterized colorimetrically (for Fe(II) and Fe(III) content) as well as by Mössbauer spectroscopy and XRD (Fig. 2). GR specimens were analyzed by SEM, a challenging undertaking as the anaerobic environment needed to be maintained rigorously to mitigate decomposition. The samples consisted of layers of flat, plate-like structures of varying shape and morphology (Fig. 3). These observations of what appear to be pure (i.e., minimal oxidation) GR samples contradict previous reports in the literature and merit further investigation.

The reliable synthesis of GR species is a key first step enabling the investigation of their redox chemistry, and we investigated GR-mediated reduction of mixed nitrate/nitrite systems. Successful efforts focused on using colorimetric methods to measure aqueous concentrations of Fe(II), Fe(III), NO₃⁻, NO₂⁻, and NH₃. These methods were adapted to a 96-well format to allow high throughput of samples analysis (Fig. 4). This system will be investigated thoroughly over the next Project year.

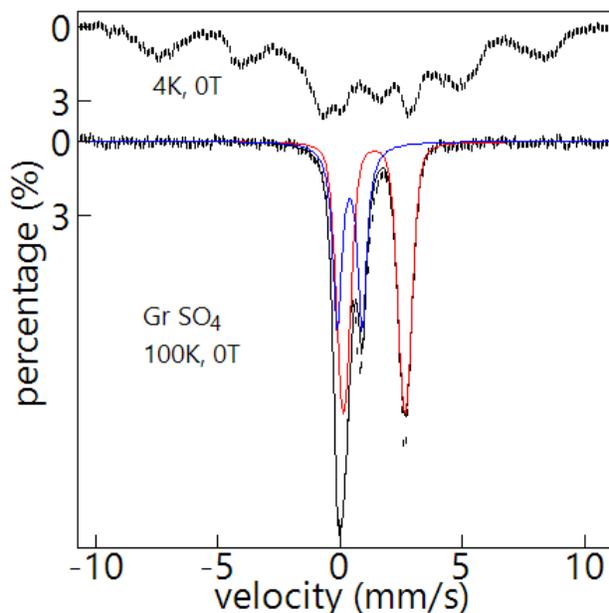


Fig. 6. Mössbauer spectra of GR₂(SO₄²⁻). The Fe(II):Fe(III) ratio was found to be 1.76. Two doublets have been identified for GR₂(SO₄²⁻) under 100 K. The isomer shift indicates one as Fe(III) and the other Fe(II). They make up 100% of the total iron component. At 4 K, both spectral components show magnetic features. The spectra were kindly provided by Yisong (Alex) Guo, Carnegie Mellon University.

We have also continued to dedicate significant effort to investigating amino acid synthesis in iron hydroxide systems, and have successfully characterized the effect of redox, pH, and temperature gradients on these reactions. These results are currently being prepared for publication. Another ongoing research effort is investigating how phosphorus (as phosphite or phosphate) reacts within iron hydroxide systems, and how amino acids affect P chemistry. In the next year we will attempt to link the GR-mediated N chemistry described above to amino acid synthesis, and characterize the effects of P on the overall system.

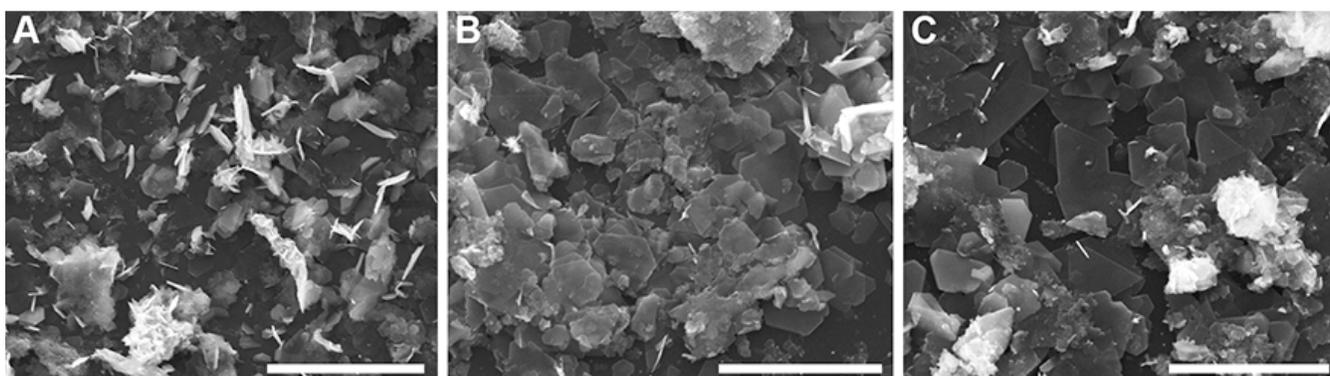


Fig. 7. Scanning electron micrographs of GR sample. A, Plates of different sizes are intermixed; scale bar, 20 μm. B, Regular and irregular structures were observed; scale bar, 10 μm. C, The plates vary from triangular to hexagonal; scale bar, 10 μm.

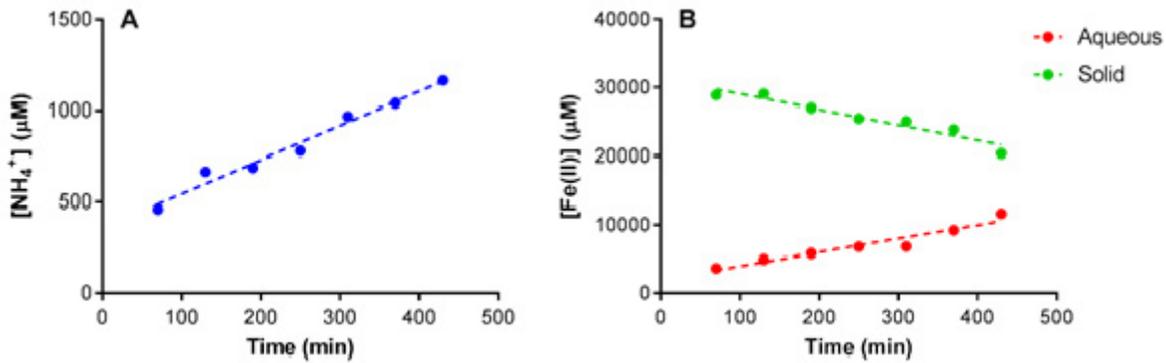


Fig. 8. Reaction of GR1(Cl⁻), Fe(II):Fe(III) ratio 3:1, with nitrate (8 mM) and nitrite (2 mM) mixture. A, Ammonia production is linear over time. B, The Fe(II) content of the GR decreases over time, while the Fe(II) concentration in solution increases.

INV 3 - Planetary Disequilibria: Characterizing Ocean Worlds and Implications for Habitability

Our major thrust in INV 3 is to explore how ocean composition affects habitability of icy bodies in our solar system and beyond. In year 3, we built on prior investigations of the radial structure and habitability of icy ocean worlds to examine how geophysical investigations by future spacecraft missions can yield new information about habitability (Vance et al. 2017a,b). The work focuses on the case for seismology as the best way to reveal detailed interior structure, composition, and present day-activity. However, the work makes a larger point about investigating habitability in worlds where the most likely place to find life may be 10s to 100s of km below the surface. It looks at how the details of the ocean's thermodynamics and the properties of the underlying rock provide more unique signatures when combined: density, ice thickness, ice-ocean interface temperature, electrical conductivity, and sound speed. This new work (Vance 2017, Vance et al., 2017b) is a foundation on which we can build investigations of the oceans' compositional evolution

via water rock interactions, the associated redox fluxes, and measurable signals at the ice surfaces and in the surrounding exospheres.

We continued to improve our analytical model by describing previously unconsidered ice-ocean interactions in Europa and other satellites (Zhu et al., 2017). This model explores the implications of variations in ice thickness for overturning circulation.

We also examined the likely porosity of Europa's near surface (Aglyamov et al. 2017) Our finding is that previous studies had overestimated the likely porosity which has important implications for the nature of heat and material transport through the ice, and thus for the internal thermal state, and for the means by which oxidants and nutrients are exchanged between the surface and the underlying ice and ocean. Our INV 3 team also continued their innovative work examining the behavior of fluids at very high pressures found in the larger icy satellites. We began measuring the phase equilibrium of high-pressure ices (III, V, VI, VII) in the presence of salts. This work complements the acquisition of a rich set

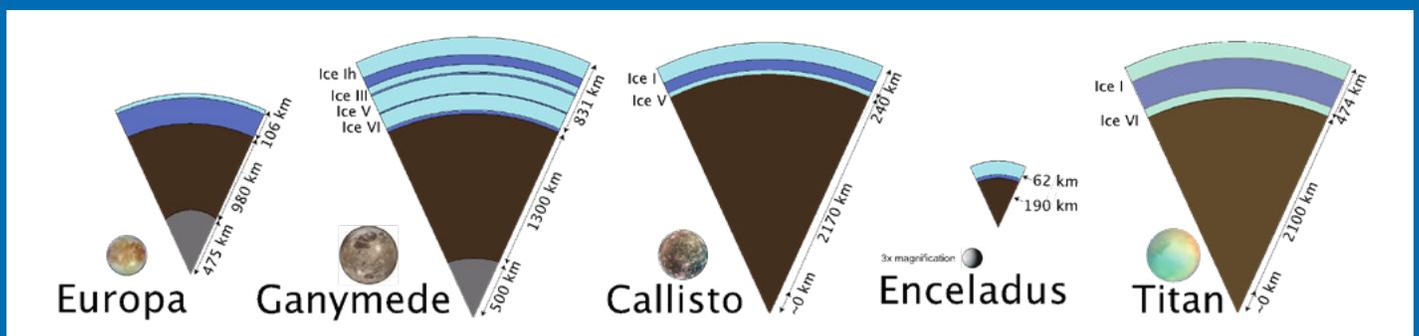


Fig. 9. Schematics showing likely internal structures for known icy ocean worlds, based on available spacecraft data, and accounting for the influences of the thermodynamics of the constituent materials, including salts and ammonia in the oceans (Vance et al. 2017b).

of thermodynamic data, based on sound speeds measured to the unprecedented accuracy of 1 part in 10,000, for aqueous solutions (NaCl, Na₂SO₄, MgSO₄, MgCl₂). The new data set covers the nearly the entire range of fluid stability relevant to ocean worlds (Fig. 5).

We also completed two studies (Choblet et al., 2017; Kalousova et al., 2017). The first numerical study investigated the amount of tidal dissipation that can be produced in the porous silicate core of Enceladus. The study shows that the amount of tidal heat can be several 10s of GW, which can explain how an ocean can persist during billions of years in such a small body. Water transport in the tidally heated permeable core results in hot narrow upwellings with temperatures exceeding 363 K, characterized by powerful (1–5 GW) hotspots at the seafloor, particularly at the south pole (Fig. 6). The release of heat in narrow regions favors intense interaction between water and rock, and the transport of hydrothermal products from the core to the plume sources. These results explain the main global characteristics of Enceladus: global ocean, strong dissipation, reduced ice-shell thickness at the south pole and seafloor activity. This endogenic activity can be sustained for tens of millions to billions of years.

The second study deals with the dynamics of the high-pressure ice layer predicted to exist in Ganymede’s interior between the ocean and the silicate core. Using a state-of-the-art convection code for two-phase fluids (solid ice and liquid water), the study demonstrates a temperate layer (porous ice with liquid water) is present at the interface with the silicates as long as the Rayleigh number, a measure of the vigor of convection in the HP ice layer, is small. The fluids generated at this interface can be mobilized and transported to the ocean by upwelling plumes. As the planet cools down and the thickness of the HP ice layer increases, the Rayleigh number increases and no more melt forms at the interface with the silicate layer, limiting the exchange between the silicates and the ocean.

As a part of our INV 3 objectives, we sought to connect our investigations of ocean worlds in the solar system with possible habitable ice-covered exoplanets (HICEPS) that probably exist in the current lists of exoworlds and the likely detection and possible interior structures of such worlds (Barnes et al. 2017).

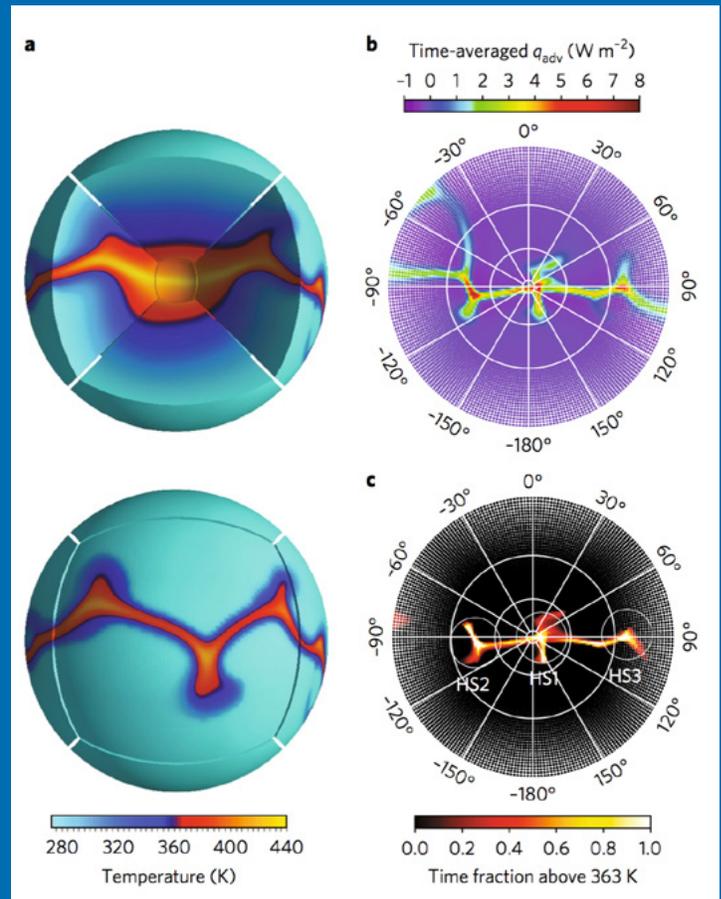


Fig. 10. Predicted temperatures (a) in the rocky portion of Enceladus due to the influence of fluids flowing through pores in the rock. The corresponding heat flow (b) and regions where temperatures persistently are near the boiling point of water (c) are also shown. (Choblet et al. 2017).

INV 4—Observable Chemical Signature on Icy Worlds: A Window into the Habitability of Subsurface Oceans

A key focus of this investigation has been to study the chemistry of frozen brines representing putative ocean composition with the goal of placing constraints of subsurface liquid composition of Ocean Worlds based on observed surface chemistry. Building on our previous work, we took a simplified model of a four-ionic component (Na, Mg, SO_4 , Cl) European ocean and mapped out what minerals form upon freezing as a function of relative ionic concentration, pH, etc. A 'flow-chart' of the freezing sequence was developed based on both published and recently acquired experimental results (Fig. 7). In performing this exercise, we began to make meaningful links between observations of the surface chemistry and the chemical environment of the internal ocean.

Prominent bright spots observed across Ceres' surface containing natrite (Na_2CO_3) and smaller amounts of NH_4Cl or NH_4HCO_3 have been proposed to originate from an internal reservoir, where subsurface brines freeze upon reaching the surface. Kinetically frozen

solutions containing the likely constituents of Ceres' subsurface brines (ammonium, sodium, carbonate, and chloride ions) were studied via infrared and micro-Raman spectroscopy, where the flash-frozen mixtures were found to preferentially form ammonium chloride and ammonium bicarbonate, even in sodium-dominated solutions. Additionally, sodium chloride only formed when sodium or chloride (or both) were present in excess in the brine solutions. Raman spectroscopy was further employed to analyze the effect of vacuum exposure on these frozen brines over longer periods of time to simulate the surface conditions of Ceres. Such vacuum exposure was found not affect speciation. This work showed that, under fast freezing conditions, ammonium bearing materials are kinetically favored. Further it suggests that the bright spots in Occator crater originate from subsurface brines mobilized by a cryovolcanic process or by the crater forming impact.

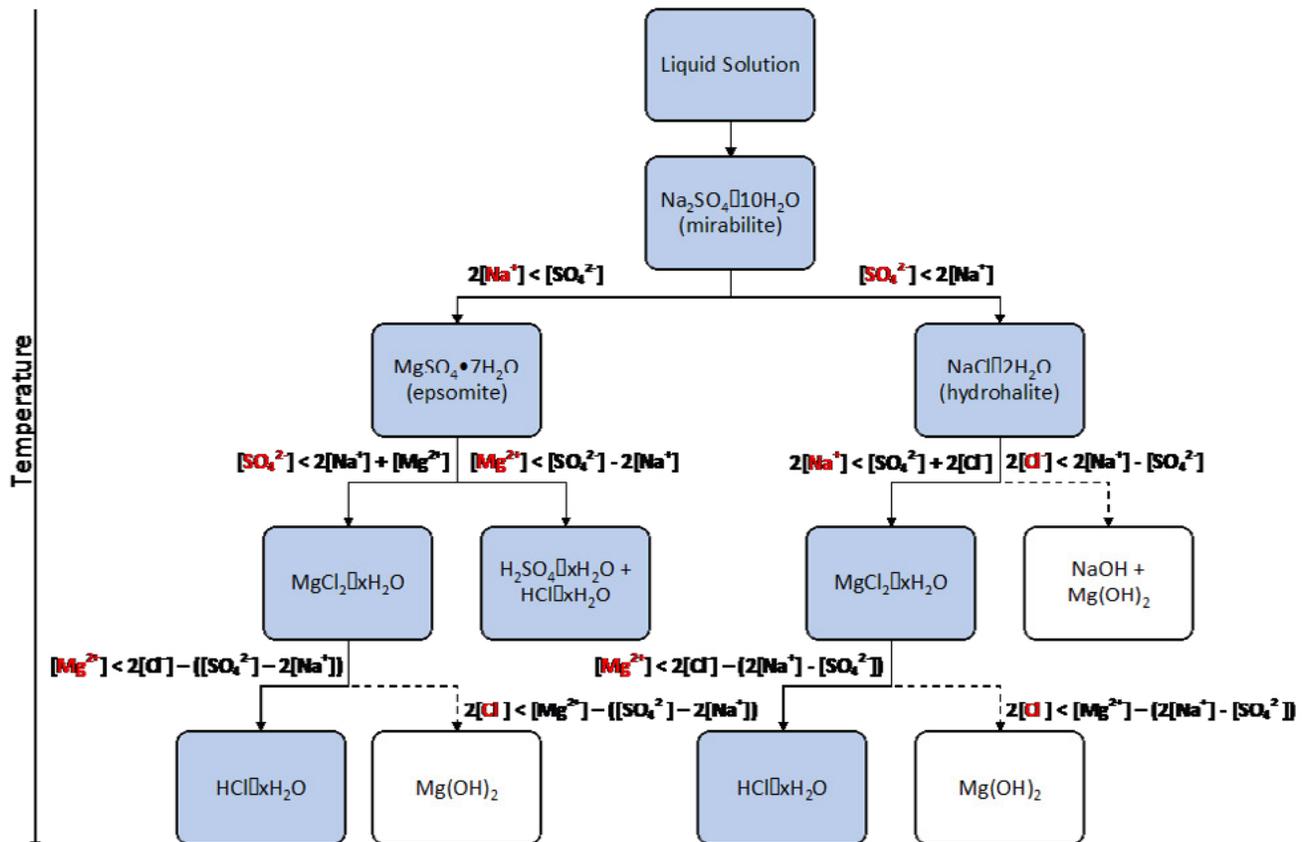


Fig. 11. Flowchart of the mineral crystallization sequence for an acidic brine containing Na^+ , Cl^- , Mg^{2+} , and SO_4^{2-} ions with decreasing temperature as a function of relative ionic concentrations. The branches in the flowchart are labeled with concentration conditions for which a given branch is followed. At each decision point, the reaction limiting ion is in red text. Solid arrows indicate experimentally verified pathways. Dotted arrows indicate paths that cannot proceed in an acidic solution. (from Johnson et al. 2018 in preparation)

Team Members

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Fig. 12. Artist's impression of Enceladus where several "lost-city" like moderate temperature hydrothermal activities on its liquid-water ocean floor and several kilometer-thick ice crust covering the ocean are shown. Credit: NASA/ESA/K. Retherford/SWRI