Subcritical phase separation, extreme arsenic enrichment, and the search for arsenotrophs in the marine shallow-water hydrothermal vents off Milos Island, Greece

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Recent investigations [1] revealed the highest arsenic concentrations measured to date for any submarine hydrothermal vent fluid, occurring in the shallow waters (<20 m) of Paleochori and Spathi Bay, off the southeast coast of Milos Island, Greece. Concentrations reached as high as 79 μM, which equals nearly 3000 times the concentration of seawater As and far exceeds the concentrations typically found in mid-ocean ridge (MOR) and back-arc basin (BAB) fluids (typically >10 μM). Orange hydrotheral precipitates at the site were characterized as non-crystalline orpiment. Both a high Cl (enriched in Cl by up to 47% compared to seawater) and a low Cl fluid (depleted by up to 66%) were encountered, and geochemical data indicate that the hydrothermal fluids underwent subcritical phase separation (boiling) and vapor/brine segregation prior to discharge.

We utilized both culture-dependent and molecular microbiological approaches to search for arsenotrophs in white and orange mats found on the seafloor in the area of venting. Field inoculations were performed using hydrothermal fluids and sediment slurries on designer growth media, including autotrophic and heterotrophic arsenate (AsV) reduction, as well as aerobic and anarobic arsenite (AsIII) oxidation, and incubated at 50 and 80 °C. Thusfar, enrichments in autotrophic AsV reduction with H_2S as the electron donor were successful, and efforts to characterize this community continue.

16S rRNA analysis showed a high diversity in the bacterial community, while archaea constituted only a minor fraction of the microbial community, with low diversity. The presence of diverse genes for the large subunit of arsenic oxidase (aroA) in the white and orange mats at Milos indicates the capability of the microbial community to metabolise arsenic.

[1] Price, R.E. Pichler, T. Planer-Friedrich, B. Bühring, S.I. Savov, I. (2011) The behavior of arsenic in subcritically phase-separated hydrothermal fluids from the marine shallow-water hydrothermal vents off Milos Island, Aegean arc, Greece. Submitted.

Interfacial tension, metastability, and solubility of solid solutions

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The existence of a relationship between metastability and solubility is widely recognized in the crystal growth literature [1]. In general, the lower the solubility of a substance the higher is its 'ability' to form supersaturated solutions. The metastability limit of a supersaturated solution can be defined by the threshold below which the solution can remain supersaturated longer than an arbitrarily chosen waiting time. This definition is in the framework of the classical nucleation theory (CNT), which connects metastability, interfacial tension (σ) and solubility. The nucleation rate depends strongly on σ, which is in turn related to the solubility by the rule that the higher the solubility, the lower σ. In CNT, the interfacial tension is actually an 'artificial' parameter, but empirical estimations [2] of σ are typically used to account for precipitation in natural systems [3] and to model the precipitation in sequential order of lesser and lesser soluble minerals constituted of the same elements, according the Ostwald step rule. In a similar way, when solid-solutions crystallize from supersaturated solutions, the partitioning of the substituting ions usually differs from the equilibrium values [4]. 'More soluble' solid solution compositions are kinetically favored and tend to nucleate even though the aqueous solution is less supersaturated for these compositions than for less soluble members. This effect has been modeled by considering the differences between the interfacial tensions of the end-members [5], but empirical estimations of σ for intermediate members of solid solutions are rare.

The aim of this work was to determine interfacial tensions of intermediate members of a number of binary (ideal and non-ideal) solid solutions and to correlate them with the corresponding solubilities (Lippmann's solidus relationships). The interfacial tensions were determined from nucleation experiments carried out at 25 °C in a solution calorimeter. As a final outcome, for each solid solution system an empirical $\sigma(x)$ function of composition is proposed.

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