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Real-time imaging of pH gradients and solution flows through silica garden membranes during tube growth

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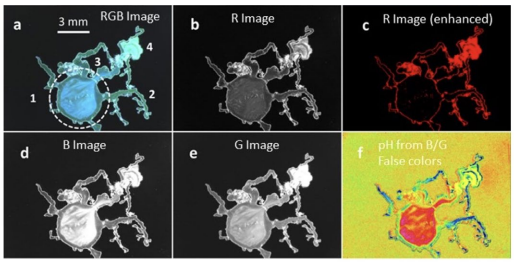

Figure 1. a) Image of the silica garden acquired with an RGB camera 10 seconds after mixing the reactants. Blue-green coloration is due to the fluorescence of the pH probe, and it is pH-dependent. b) red component of the image, this is essentially pH-independent and shows the morphology of the membrane. The contrast of the red image was enhanced with ImageJ to get a better-defined membrane profile, as shown in c. The blue and green components are shown in d and e, respectively, and they were used to calculate the intensity ratio R and hence the pH map shown in f.

Chemical gardens are self-assembled tubular precipitates formed by a combination of osmosis, buoyancy, and chemical reaction [1]. In many cases, the tube wall is a bilayer structure with the properties of a diaphragm and/or a membrane. The interest in silica gardens as microreactors for materials science has increased over the past decade because of their ability to create long-lasting electrochemical potential [2]. This is why they can catalyze prebiotic condensation reactions [3]. Understanding and controlling the microscopic processes involved in forming and developing the inorganic membranes constituting chemical gardens is important in mineral self-organization and chemistry of materials, as well as in understanding the origin of life on Earth with implications in primitive life detection. Exploiting the combination of different simultaneous optical imaging techniques, we achieved an unprecedented complete overview of the processes involved in silica garden formation and growth. In particular, membrane formation and growth are visualized in real-time at the macroscopic and microscopic levels in a quasi-2D regime. Moreover, the unusual behavior of conventional pH probes in the saturated metal solution is investigated in detail and exploited to perform ratiometric pH mapping in real time either in the low magnification range, with a color camera, or at high magnification, with a confocal microscope. The circular membrane initially formed in the experiment contains an acidic metal-saturated solution with a pH considerably increased near the interface. Because of this alkalinization process, branches initially formed, erupting from the main circular membrane and showing a pH of about two units higher with respect to the starting metal solution. The pH drop also leads to the formation of microparticles that can be optically tracked to visualize the flow of the solution. Using the same probes and setup, confocal fluorescence microscopy allowed us to map pH in the microtubes with higher resolution by ratiometric analysis.

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