Spiral patterns and morphology in chemical systems

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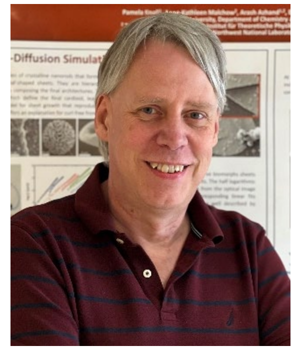


Synopsis

This talk will showcase different examples of chemical systems undergoing self-organization far from the thermodynamic equilibrium. Starting with a brief look at the history of chemical waves and the work of the field’s pioneers, we will discuss similarities to the growth of unusual polycrystalline solids called biomorphs and go beyond to the self-propulsion of chemical garden tubes.

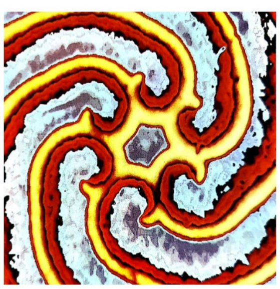
Abstract

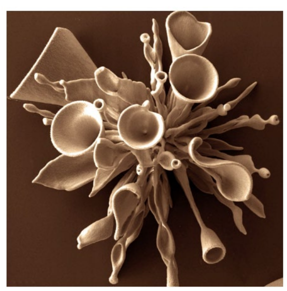
Living systems masterfully program molecular events to create macroscopic shapes and dynamics. Can the same be done with inorganic chemistry? I believe the answer is yes if reactions are kept far from the thermodynamic equilibrium (and your expectations are not too high). In my talk, I will introduce several self-organizing chemical systems including the Belousov-Zhabotinsky (BZ) reaction, chemical gardens, biomorphs, and self-propelled catalytic units capable of swarm-like cooperativity. These examples will illustrate unexpected behaviors if reactions are allowed to unfold in space and time. Perhaps not surprisingly, similar processes are used by living systems suggesting system-level interpretations of important biological phenomena (e.g. cardiac arrhythmia) and novel approaches to materials science and engineering.

Our discussion of the BZ reaction will focus on three-dimensional patterns and discuss them in the context of pioneering work by Profs. Stefan Müller, Benno Hess, and Art Winfree. These and other prominent scientists studied two-dimensional spirals, the basic building blocks of scroll waves that provide the foundation for understanding 3D dynamics. While spiral waves rotate around point singularities, scroll waves rotate around 1D filaments that move according to their local curvature. Today tomographic methods, numerical simulations, and controlled manipulations are used to obtain deeper insights into the local pinning to heterogeneities as well as scroll behavior in gradients. Many of these studies, including the results to be presented, are motivated by similar phenomena in the ventricles of the human heart and their link to sudden cardiac death.

Similar nonlinear wave dynamics appear to rule the growth of certain polycrystalline assemblies known as biomorphs. The smoothly curved and life-like biomorphs form when aqueous solutions of $BaCl_2$ and silicate react with CO_2. The shapes are about 100 µm in size and include helices, funnels, and urns. The entirely abiotic biomorphs can form from natural spring waters and constitute intriguing problems for the identification of Earth's earliest microfossils as well as the search for life's remnants on Mars and other planetary bodies.

Lastly, we will present novel results on chemical gardens (CG). In the classical CG experiment, these colorful, hollow precipitate tubes form when a salt crystal is placed into an alkaline sodium silicate solution. Specific aspects to be covered include a cellular automaton model of CGs, the involvement of colloidal particles in their growth, and applications as self-propelled micro-rockets. Our model considers, CG growth based on a simple A+B->C reaction and a lattice for which each site is exactly one of these three species. We show that the aging of the material modulates the likelihood of mechanical changes in the position of the solid product C and that a strong preference for changes in the fresh material causes filament (2D) and tube (3D) growth. The self-propulsion of CG tubes is demonstrated for Mn-containing units in H_2O_2 solutions. The tubes are propelled forward by the rhythmic ejection of O_2 microbubbles formed in the catalytic tube cavity. We also observed a characteristic acoustic signature of this propulsion that allows detailed analyses of the bubble dynamics. These analyses reveal that the tube speed is determined by the tube diameter (which approximates the step size per bubble) and the bubble ejection frequency (which is nearly proportional to the H_2O_2 concentration). Lastly, we will discuss the swarming behavior of the CG tubes and their interactions with bubble rafts.





References

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