2017 International Bionic Innovation Competition

Assemblies of Graphene Quantum Dots: From Hollow Nanoshells to Protocells for Life Origin

The key step in understanding the genesis of life on Earth, and potentially other planets, is the appearance of the first metabolic compartments, also known as protocells. Recently, we found that the graphene quantum dots (GQDs) spontaneously form hollow nanoshells in the presence of Earth-abundant metal ions in water. The GQD nanoshells represent the missing link between the inorganic world and the appearance of the first cells. GQDs and their assemblies have the ability to (a) serve as early prebiotic compartments for preliminary metabolism, (b) catalyze the formation of complex organic molecules, and (c) propagate by self-replicating. The mechanism was studied using a combination of experiments and computations, including state-of-art electron microscopic techniques and molecular dynamic simulations.

The GQD nanoshells were formed through coordination bonds between carboxyl groups on GQD edges and Earth-abundant metal ions such as Fe^{+3}, Zn^{+2}, and Ni^{+2}. GQD nanoshells represent a type of supraparticles that form a shell-like structure due to the balance of electrostatic repulsion and coordination bonds connecting their edges. The nanoshells show some structural similarities with the lipid membranes, such as high surface charge, carbonaceous building blocks, and combination of hydrophilic and hydrophobic regions。

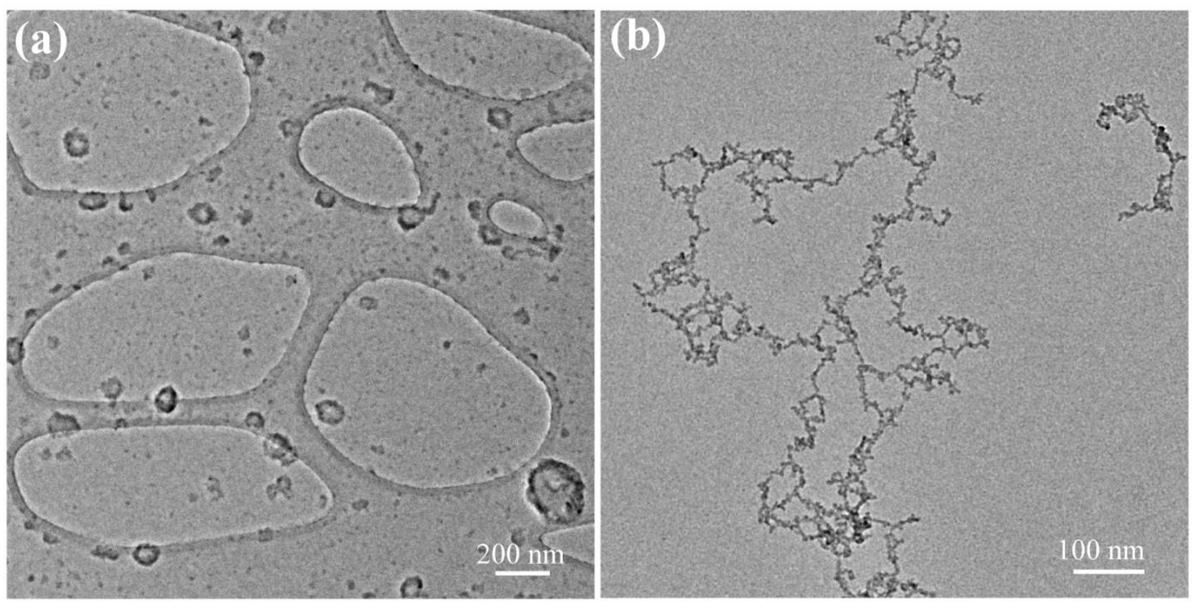

Figure 1. Self-assembly of GQDs with metal ions. (a) nanoshells and (b) nanochains.

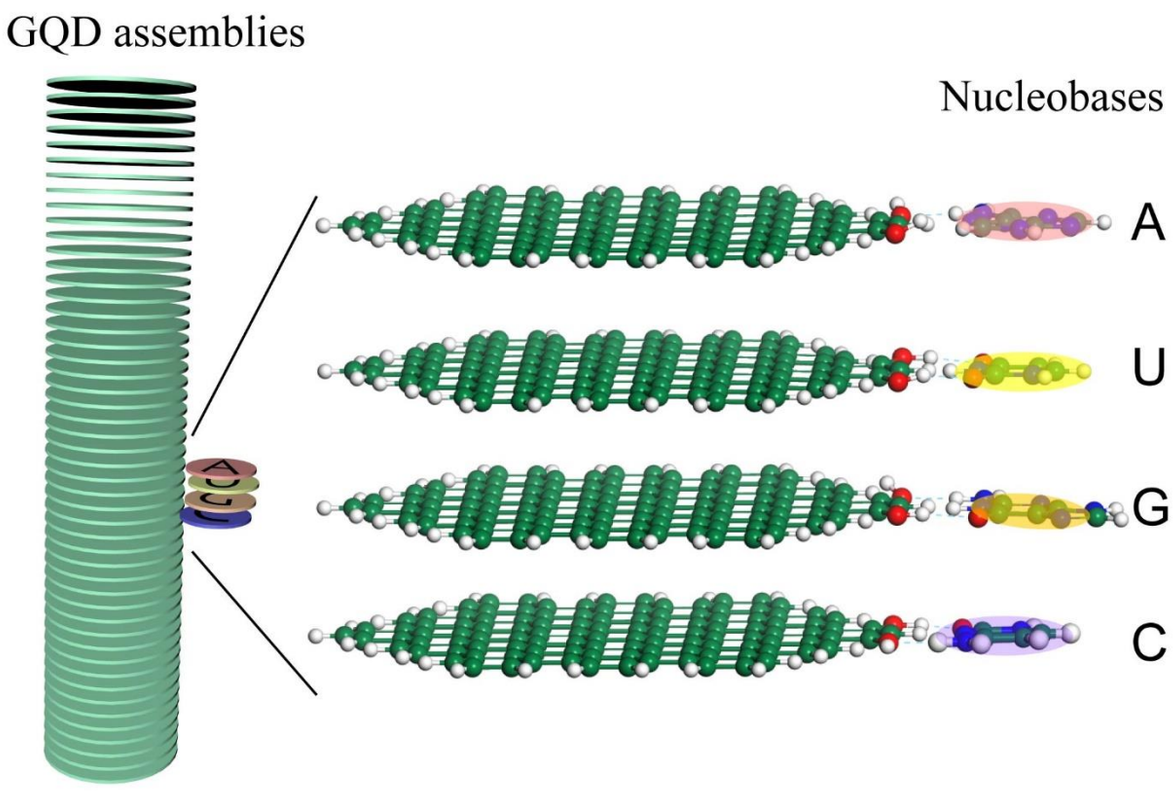

Figure 2. RNA synthesis templated by GQD assemblies.

Discussion

Cells contain complicated organelles and thousands of enzymes to propagate complex metabolism. When life first appeared on Earth, there should be a simpler form of cells, namely protocells. Scientists do not know a lot of details about protocells, as their direct descendants became extinct long ago in evolution without any traces. Protocells are thought to be composed of a membrane and metabolic machinery necessary to replicate themselves. What appeared first–the membrane or the biomolecular machinery–is a chicken-and-egg question that we are addressing in this project.

Tiny graphene segments were a part of the primordial environment on Earth. We found that these inorganic graphene nanosheets can behave like lipids, the building blocks of cellular membranes, and spontaneously assemble into compartments called nanoshells. These nanoshells could have served as the early membranes that protected the metabolic machinery represented by early enzymes. The abundance of graphene segments in space makes them very promising to be the building blocks of early protocells on prebiotic Earth. Our project will help to understand how graphene-based “protocells” evolved from the inorganic world and will elucidate their relationship to modern cells in structural properties and functions.

- Paper: Coordination assemblies of graphene quantum dots. *Nature Chem.* Under Review.