Chromophore amphiphile-polyelectrolyte hybrid hydrogels for photocatalytic hydrogen production

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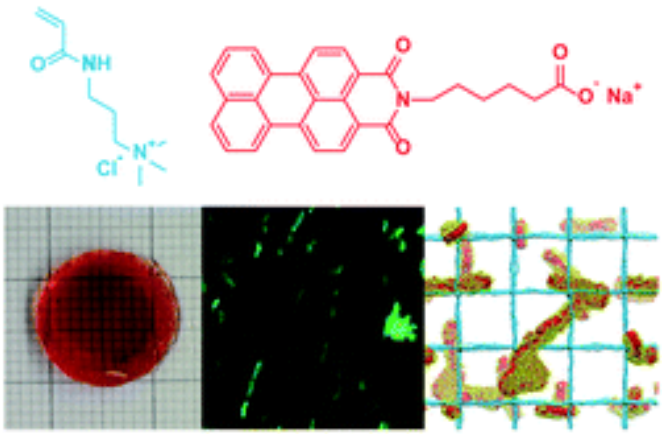
We have demonstrated the possibility of entrapping supramolecular light harvesting assemblies for photocatalytic reactions within crosslinked polyelectrolytic hydrogels over very long-time scales. This entrapment is possible by triggering self-assembly of chromophore amphiphile monomers through organic to water solvent exchange within the covalent network. The retention mechanism of the crystalline chromophore amphiphile assemblies in an aqueous environment results from both electrostatic interactions and physical entrapment in the mesh size of the network. **Interestingly, coarse grained molecular dynamics simulations suggest that chromophore assemblies are nucleated at the nodes of the three-dimensional covalent network.** Proof-of-principle experiments on photocatalysis of hydrogen production showed that optimized systems could be reused in multiple cycles of photocatalysis after refreshing the system for removal and addition of small molecule components.



Covalent hydrogel + chromophore nanoribbons:
Reusable photoabsorber for hydrogen production

Significance and Impact

The trapping mechanism strategy of light harvesting assemblies would lend itself to compartmentalization of different chemical reactions in neighboring hydrogels with chromophore amphiphiles of different energy levels for multistep synthesis, as well as possibilities for using stimuli-responsive hydrogels as the covalent network to control the photocatalytic reactions.

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