Mimicking the antenna system of green plants by supramolecular organization of dyes in nanochannels

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Natural photosynthesis is the essential process for life on earth. But its overall thermodynamic efficiency for the production of fuel is low and depends much on optimal soil, temperature, and humidity conditions. Natural photosynthesis has many other important tasks to fulfill than just conversion of solar light into a chemical fuel. A long-standing challenge has therefore been the development of an artificial photosynthetic system that is specialized on the energy conversion process.[1]

Green plants have developed sophisticated and highly efficient tools for harvesting light and transporting electronic excitation energy. Their antennas consist of regular arrangements of chlorophyll molecules held at fixed positions by proteins. Light absorbed by any of these chromophores is transported to the reaction center, providing the energy necessary for the chemical processes to be initiated. A green leaf consists of millions of such well organized antenna devices. Mimicking the light harvesting properties of the natural antenna has been an old dream and challenge of photo chemists. Different strategies for achieving this goal have been followed.[2]

We [3-5] and others [6] have focused on a design based on the incorporation of chromophores into the one-dimensional (1D) channels of zeolite L (ZL) and by organizing the composites in specific ways. ZL was found to be an excellent host for the supramolecular organization of many different molecules and complexes. The range of possibilities for filling its 1D-channels with suitable guests is much larger than one might expect.[4] Constraints imposed by the host lead to supramolecular organization of the guests. The arrangement of dyes inside the ZL channels is what we call the first stage of organization. It allows light harvesting within the volume of a dye-loaded ZL crystal and the radiationless transport of electronic excitation energy to a well positioned acceptor. The second stage of organization is realized by coupling either an external acceptor or donor stopcock fluorophore at the ends of the ZL channels, which can then trap or inject electronic excitation energy. The third stage of organization is obtained by interfacing the composites to an external device via a stopcock intermediate. A possibility to achieve higher levels of organization is by assembling the composites into ordered structures. The usually strong light scattering of ZL can be suppressed by refractive index matching and avoidance of micro phase separation.

Our understanding of guest-host interactions and the interpretation of experimental results has much improved by performing advanced theoretical modeling studies.[6,7]

Concepts, results, and possible applications are discussed.

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