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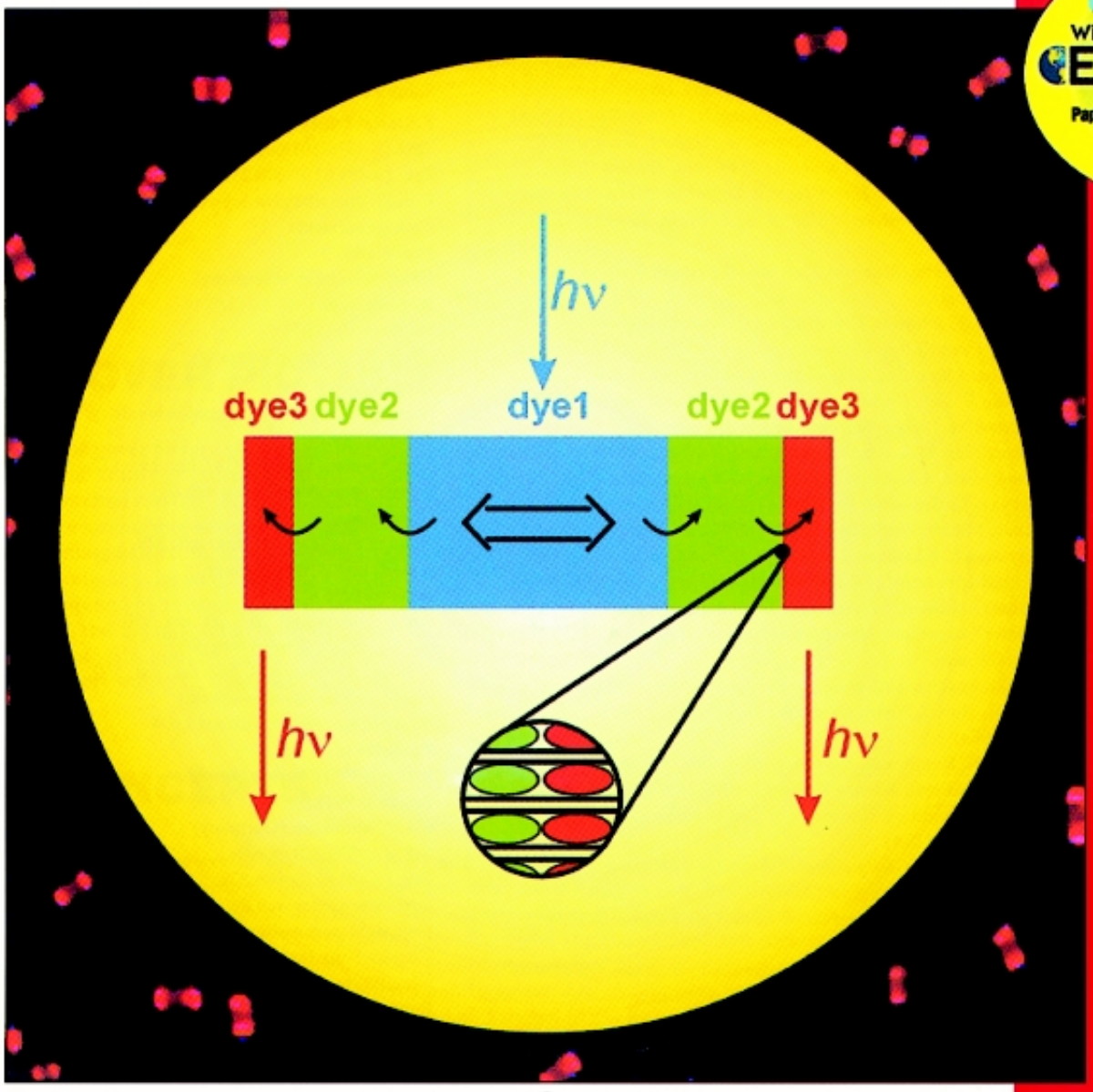

Concepts

A Library of Carbohydrate Mimics and Polyketide Segments for the Synthesis of Marine Natural Products

H. M. R. Hoffmann and A. M. Misske

Multi-Component Transformations Lead the Way

H. Bienaymé et al.

WILEY-VCH

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Cover Picture

G. Calzaferri et al.

The cover picture shows a schematic view of a bipolar antenna for light harvesting and transport surrounded by fluorescence microscopy pictures of individual antenna crystals. Zeolite L microcrystals of cylinder morphology are used as host for organising several thousand dyes as monomers into well defined zones. The enlarged section shows the organisation of individual dye molecules at the domain boundary between dye2 and dye3. The microscopy pictures demonstrate the antenna behaviour: They show the red fluorescence of dye3, located at both ends of the crystals, after selective excitation of the blue dye1 in the middle. The synthesis and characterization of these dye loaded zeolite L sandwiches are described by G. Calzaferri et al. on p. 3456ff.

Dye-Loaded Zeolite L Sandwiches as Artificial Antenna Systems for Light Transport

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Abstract: The synthesis and characterization of dye loaded zeolite L sandwiches acting as artificial antenna systems for light harvesting and transport is reported. A set of experimental tools for the preparation of neutral dye-zeolite L materials ranging from low to maximum packing densities has been developed. The role of co-adsorbed water and the distribution of molecules between the inner and the outer surface were found to be the determining parameters. *p*-Terphenyl (*p*TP) turned out to be very suitable for studying these and other relevant parameters of neutral dye-zeolite L materials. We observed that *p*TP located in the channels of zeolite L can reversibly be displaced by water. This can be used when synthesizing such materials. We also observed that all-*trans*-1,6-diphenyl-1,3,5-hexatriene (DPH) which is very photolabile in solution is stable after insertion into zeolite L. By combining our extensive knowledge of these systems with ion-exchange procedures developed in an earlier study, we have realized the first bi-directional three-dye antenna. In this material the near UV absorbing compounds DPH or 1,2-bis-(5-methyl-benzoxazol-2-yl)-ethene (MBOXE) are located in the middle part of zeolite L nanocrystals followed on both sides by pyronine (Py) and then by oxonine (Ox) as acceptors. Fluorescence of the oxonine located at both ends of the cylindrical zeolite L crystals was observed upon excitation of the near UV absorber in the middle section at 353 nm, where neither oxonine nor pyronine absorb a significant amount of the excitation light.

Keywords: artificial antenna · energy transfer · host–guest chemistry · supramolecular chemistry · zeolites

Introduction

The structural, morphological, physical, and chemical variety of zeolites has led to applications in different fields such as catalysis,[1] ion exchange, membranes,[2, 3] and chemical sensors where dynamic processes involving ions or adsorbate molecules play an important role. Situations where the zeolites mainly serve as host for supramolecular organization of molecules, ions, complexes, and clusters to prepare materials with new properties such as nonlinear optical,[4] quantum-size,[5, 6] micro laser,[7] and artificial antenna characteristics[8–10] are new fields of growing interest.[11–13] Some of these new materials can be considered as a static and stable arrangement of guests in the zeolite host under a broad range of conditions.[14] In other cases, the adsorption, desorption or ion exchange of molecules or ions are reversible processes which lead to a wide range of phenomena.[15–17]

Our current interest in zeolites is focused on their use as host materials for different kinds of supramolecular organization.[6, 8] This article concentrates on the synthesis of dye-loaded zeolite L sandwiches acting as artificial antenna systems. It has been demonstrated both theoretically and experimentally that by organizing cationic dyes in the one-dimensional channels of zeolite L nanocrystals an artificial antenna system for light harvesting and fast anisotropic energy transport can be realized.[9, 10, 18, 19] In this material two different kinds of dyes with appropriate photophysical properties are organized as monomers in spatially separated domains. The structure of the system allows selective excitation of the central part and fast transport of the energy along the channel axis to the top and bottom faces of the crystals. Pyronine and oxonine have been used as chromophores in this material. Blue-green light is selectively absorbed by the pyronine molecules located in the middle part of the crystals with cylindrical morphology. The energy then migrates to the crystal ends where it is captured by the oxonine and then emitted as red luminescence.

In the presented work neutral molecules absorbing UV light were incorporated in order to evaluate the light harvesting properties of this material. The resulting three-dye zeolite L sandwich materials open the door to new systems with fascinating properties. We show that a bi-directional antenna for light collection and transport can been prepared so that the whole visible light spectrum can be used. The light

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