Laboratoire PPSM – UMR CNRS 8531
Photochimie et Photophysique Supramoléculaires et Macromoléculaires

Soutenance de thèse

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«Photoswitchable hysteresis based on multichromophoric architectures»

Photoswitchable fluorescent materials have potential applications in optical information storage or super-resolution imaging area, attracting considerable research developments. In such molecular systems, competitive photoisomerization pathways and kinetics under UV and visible light irradiation, corresponding to forward and backward photoinduced reactions, are expected to open a fluorescence hysteresis cycle. With this goal in mind, we aim to generate fluorescence hysteresis effects with multichromophoric architectures based on cyclodextrin multivalent platforms bearing several dendritic linkers, substituted by a variable number of photoswitchable and/or fluorescent units. The work carried out in this PhD covers the studies on a E/Z photoisomerizable fluorophore (derived from dicyanomethylene-4H-pyran, DCM), multichromophoric systems holding multiple DCM units (multi-DCMs), or a combination of DCM and another photochromic unit (diarylethene, DAE). After a general review on molecular photoswitches (Chapter I), the DCM itself has been investigated by spectroscopic techniques (steady state, time-resolved) and theoretical approaches (Spin-Flip Density Functional Theory, SF-DFT) to unravel the fluorescence and photoisomerization processes, especially through its conical intersections (Chapter II). In Chapter III, several photoisomerizable and fluorescent DCM have been then combined together by click chemistry strategies into dendritic molecules (multi-DCMs) and their spectroscopic studies indicate effective intramolecular Förster resonance energy transfer (FRET) processes. Fluorescence anisotropy and molecular dynamics simulations have been carried out to confirm and support the FRET models in such multi-DCMs. In the end, the Chapter IV describes our efforts to synthesize a DCM-DAE combined architecture, leading to a “trichromophore” system (1 DCM, 2 DAE), further assembled into 12-valent and 42-valent multichromophores. These complex multichromophoric systems show efficient intramolecular FRET pathways and demonstrate the fluorescence hysteresis effect, which depends on the number of chromophoric units. Such fluorescence hysteresis features, demonstrated by means of smart photoswitchable and fluorescent multichromophores, are very promising for a wide range of future developments, since we can envision complex multi-state optical data storage or photoactive biosystems capable to be grafted by click chemistry for enhanced fluorescence imaging, via precise control by light.

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