

Photo/Redox-Responsive 2D-Supramolecular Assembly Involving Cucurbit[8]uril and a Star-Shaped Porphyrin Tecton

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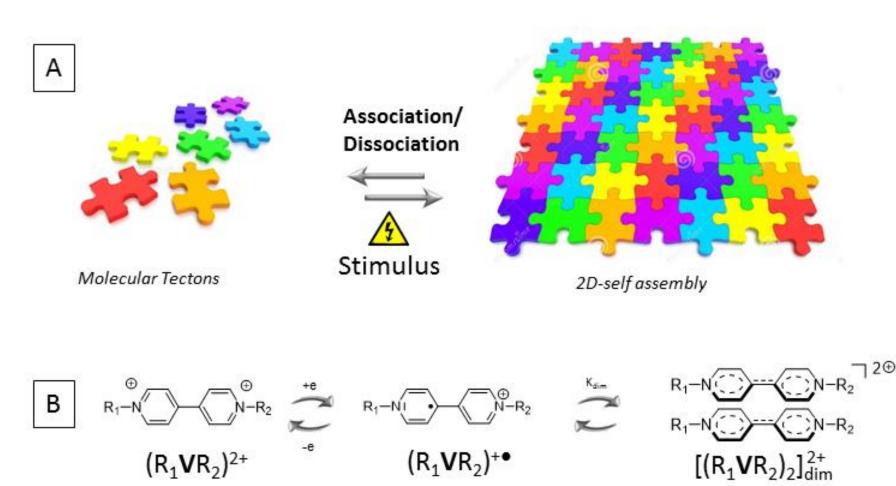
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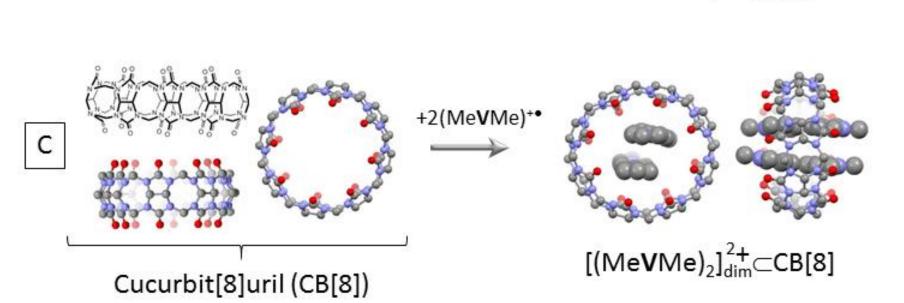
Introduction

Supramolecular assemblies whose association or organization between tectons can be controlled by external stimuli have been found to be of great interest in various areas such as molecular electronics, analytical chemistry and materials science.^[1] In this context, we are interested in (supra)molecular metamorphism involving dimerization of π -conjugated radicals as driving force. Our group has been focusing over the past few years on the development tailor-made redox-controllable molecular or supramolecular of systems involving electro-generated viologen-based π -cation radicals as key responsive and/or assembling elements, using their ability to self-assemble into sandwhich-like, dimeric entities called π -dimers. Such π -dimers can be observed in standard temperature and concentration ranges, when using barrel-shaped cavitands, known as cucurbit[8]urils, whose inner cavity is ideally suited to the inclusion of two viologen-based radicals.^[2]

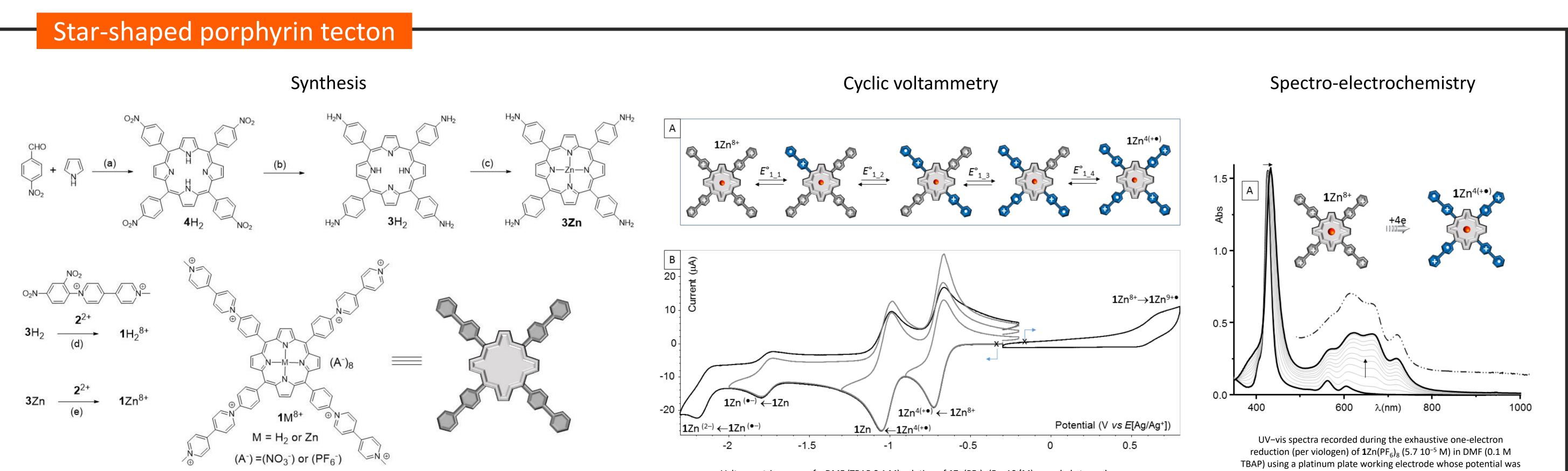


Conclusion

- Formation of discrete 4:1 pseudorotaxane-like caviplexes: 4 CB[7 or 8] / viologen-based star shaped porphyrin guest
- Formation of intermolecular π -dimers requires CB[8] acting as an assembling element (electrochemical, theoretical and spectroscopic analyses)



CB[8] hosts proved useful to promote the redox-triggered formation of a 2Dsupramolecular assembly in solution (electrochemical, chemical or photochemical stimulus)



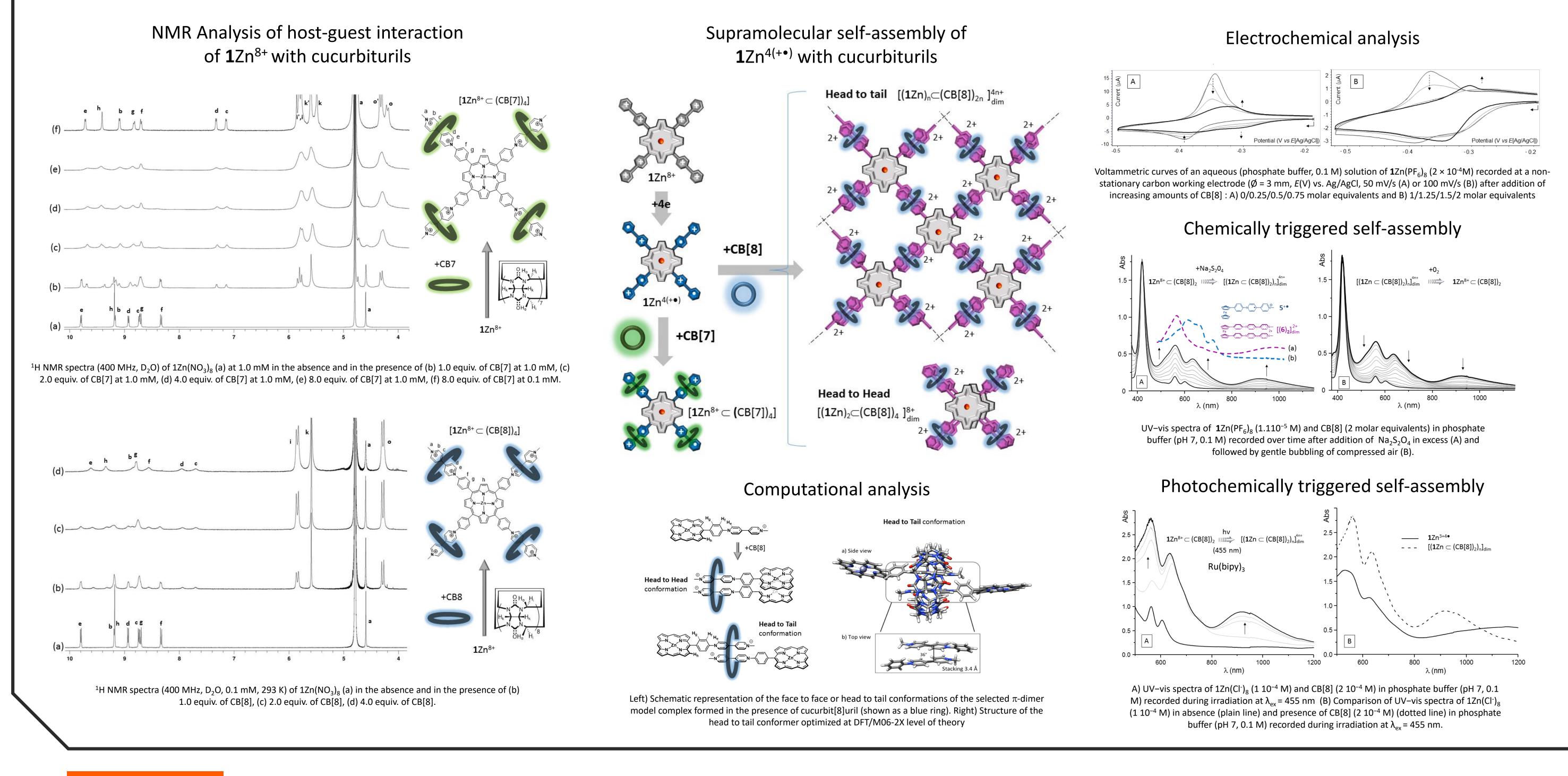
(a) EtCO₂H, Ac₂O, reflux, 4 h, 30%; (b) SnCl₂·2H₂O, conc. aq. HCl, reflux, 16 h, 62%; (c) Zn(OAc)₂, DMF, 25 °C, 30 min, >99%; (d) 2(PF₆)₂, EtOH, H₂O, reflux, 18 h, 29%; (e) 2(PF₆)₂, EtOH, H₂O, reflux, 18 h, 32%.

Voltammetric curves of a DMF (TBAP 0.1 M) solution of $1Zn(PF_6)_8$ (5 × 10⁻⁴M) recorded at a carbon working electrode (\emptyset = 3 mm, *E* vs Ag/Ag+ (10⁻²M), v = 0.1 V·s⁻¹).

fixed at $E_{ap} = -0.85$ V vs $E_{Ag/Ag+}$ (10 mL, I = 1 mm, t = ~30 min).

Supramolecular assembly involving cucurbituril

We have used this star-shaped porphyrin tecton to control the association/dissociation process of a 2D-supramolecular self-assembly with cucurbit[8] urils by means of electrochemical, chemical, and photochemical stimuli.^[3]



References

[1] X. Yan, F. Wang, B. Zheng, F. Huang, Chem. Soc. Rev. 2012, 41, 6042–6065; D. Xiang, X. Wang, C. Jia, T. Lee, X. Guo, Chem. Rev. 2016, 116, 4318–4440; I. V. Kolesnichenko, E. V. Anslyn, Chem. Soc. Rev. 2017, 46, 2385–2390; S. Kassem, T. van Leeuwen, A. S. Lubbe, M. R. Wilson, B. L. Feringa, D. A. Leigh, Chem. Soc. Rev. 2017, 46, 2592–2621; A. Wang, W. Shi, J. Huang, Y. Yan, Soft Matter 2016, 12, 337–357. [2] H. D. Correia, S. Chowdhury, A. P. Ramos, L. Guy, G. J.-F. Demets, C. Bucher, Polym. Int. 2019, 68, 572–588. [3] S. Chowdhury, Y. Nassar, L. Guy, D. Frath, F. Chevallier, E. Dumont, A. P. Ramos, G. J.-F. Demets, C. Bucher, Electrochim. Acta 2019, 316, 79–92.