A synthetic strategy based on the hydrophobic effect to build molecular interlocked structures

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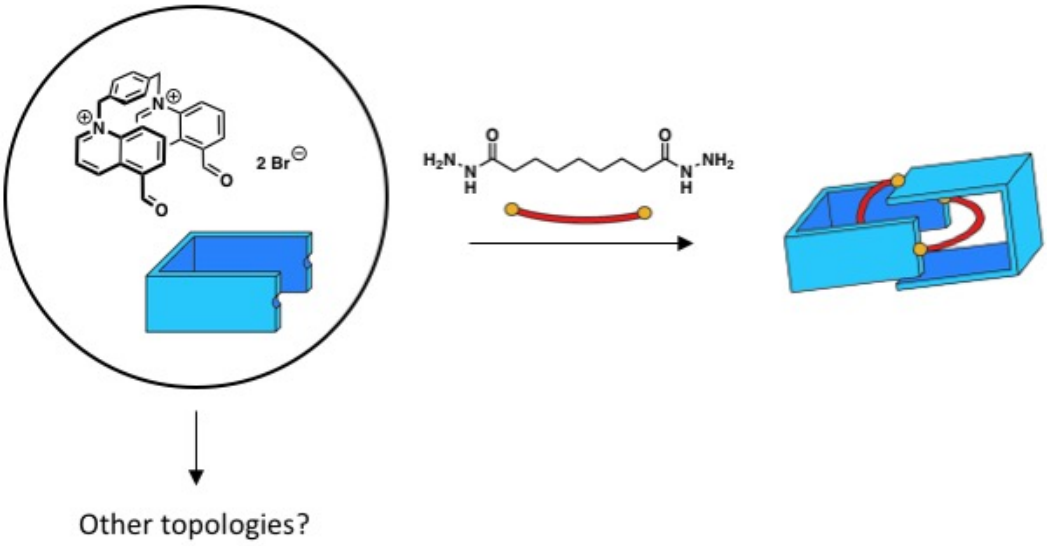
In spite of significant efforts spanned over the past 40 years, synthesising molecular links and knots more complex than [2]catenanes remains one of the biggest challenges of modern chemistry. Traditional approaches to build interlocked molecules consist in pre-organizing building blocks using metal templation, hydrogen-bonds or donor-acceptor pi-pi interactions.[1,2] The recent discovery of a trefoil knot by the Sanders group suggests that exploiting the hydrophobic effect could provide the basis of a powerful alternative strategy.[3] Inspired by this discovery, we hypothesised that hydrophobic building blocks could self-assemble in water into complex interlocked architectures depending on: 1) the size of the p-surface; 2) the amount of flexibility; and 3) the geometry of each parts. We present here a simple system where all these parameters can be varied with ease. The linkage between the building blocks is reversible (e.g. imine, hydrazine and oxime bonds). When the all these parameters are favourable, a mechanism of error-correction transforms trivial macrocycles into the thermodynamically more stable interlocked structure. We first proved the validity of our approach with the synthesis of a [2]catenane. We believe that the same system will soon allow us to build more complex topologies.



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[3] Nandhini Ponnuswamy, Fabien B. L. Cougnon, Jessica M. Clough, G. Dan Pantoş, Jeremy K. M. Sanders, Science, **2012**, 338, 783-785.