THE HONG KONG POLYTECHNIC UNIVERSITY
香港理工大學

Faculty of Science

Department of Applied Biology and Chemical Technology

Prof. Peter H. Seeberger
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Dear Prof. Seeberger,

Enclosed is our manuscript entitled "*Supramolecular Assemblies of Donor-Acceptor Stenhouse Adduct Amphiphiles as Macroscopic Soft Scaffolds*" that we would like to submit as an Article to the *Beilstein Journal of Organic Chemistry*, under thematic issue: Harnessing light energy with molecules for Young Investigator Award application.

The development of stimuli-responsive soft functional materials by molecular design is at forefront of contemporary chemical sciences with a key role played by molecular machines. The transition from static to dynamic complex functional systems brings fascinating opportunities but also major challenges in merging synthesis, responsive behavior, and supramolecular organization across length-scale from nano- to macroscopic dimensions. The control of macroscopic supramolecular transformation based on a visible-light responsive molecular machine design (instead of bio-damaging UV-light) enabling a system performing a macroscopic function, is remained largely unexplored.

Here we describe a supramolecular system that is powered by a photoswitch donor-acceptor Stenhouse adduct (DASA) and is controlled by visible-light stimulation. It allows the controlled macroscopic supramolecular transformation while being completely under control of external input signals. An additional key design feature is that the whole system operates in water. The design features an amphiphilic DASA that bears a carboxylate motif, connected with different chain length of alkyl-linker, introduced to induce aqueous solubility and maintain the propensity to self-assemble into a supramolecular assembly. The supramolecular assembly is susceptible to visible-light stimulation providing supramolecular transformation into micellar assemblies. In addition, the supramolecular organization, applying a shear flow method, allows assembly of the nanofibers into a macroscopic soft scaffold.

This macroscopic soft functional material, based on DASA amphiphiles, demonstrates that indeed complex dynamic functions can be exerted by a supramolecular system. The macroscopic soft scaffold is entirely

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based on supramolecular organization, comprising mainly water (~90%) and responsive molecules powered by visible light but not bio-damaging UV-light.

We consider these results as a major step forward in the design of artificial soft responsive materials, how to exploit molecular motion and to use the principles of supramolecular organization of small molecules to construct macroscopic materials with the whole system operating in aqueous media as an additional major challenge. We consider our results to be of major interest to the wider chemical community. We would greatly appreciate publication of this work in the *Beilstein Journal of Organic Chemistry*.

Full experimental details are included as supporting information.
Thank you very much.

With best regards,

Franco Leung

Dr. Franco King-Chi Leung
Assistant Professor