

### Repairecture

**Innovative repair concepts for polymer-based materials and components along their life-time**

Programme: COMET – Competence Centers for Excellent Technologies

Programme line: COMET-Module

Type of project: Project 1.02 - Fast repairable dynamic covalent polymer networks without creep for a circular economy of crosslinked polymer components, 2024-2027, multi-firm



## LOCAL AND REVERSIBLE SELF-FOLDING OF CROSSLINKED POLYMERS

REVERSIBLE SWITCHABLE CATALYSTS WERE DEVELOPED THAT ENABLE MOLD-FREE SELF-FOLDING OF CROSSLINKED PHOTOPOLYMERS.

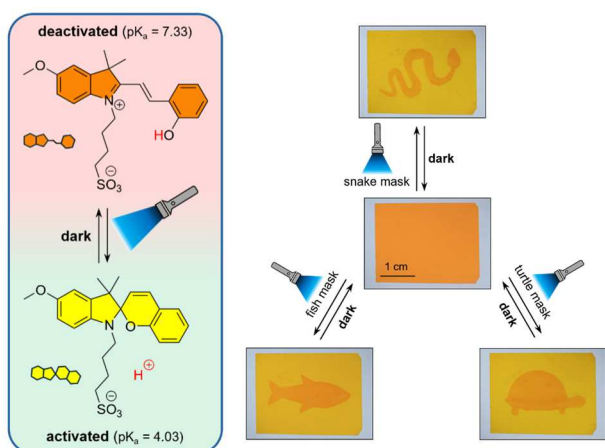
Thermosetting polymers, having a covalently cross-linked network structure, offer outstanding mechanical performance and chemical resistance. However, their robust nature poses critical challenges for the recycling, welding or reshaping of these materials. Dynamic polymer networks aim to bridge this gap between performance and sustainability by the introduction of dynamic covalent bonds capable of reorganizing the network topology in response to an external stimulus. The resulting change in viscoelasticity imparts weldability, malleability and reparability in the crosslinked polymers. In the COMET module project *Repairecture*, a merocyanine photoacid was synthesized in collaboration with

University of New South Wales and University of Melbourne. The synthesized catalyst enables fast and reversible control of viscoelastic properties in crosslinked polymers undergoing transesterification. The work was published in the renowned *Journal of the American Chemical Society*, JACS. (<https://doi.org/10.1021/jacs.5c13227>).

### Impact and effects

When exposed to visible light, the photoacid's activated spiropyran form enables a macroscopic material flow through acid-catalyzed transesterification. Switching off the light yields the

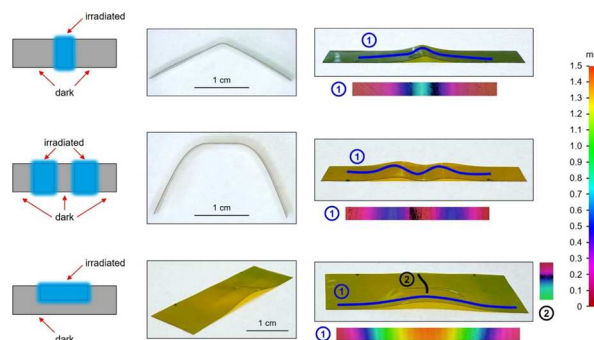
deactivated merocyanine form and a creep-resistant thermoset is obtained. Stress relaxation experiments clearly demonstrate a remarkable difference in mechanical properties resulting from the photoacid's isomerization state. Compared to previously studied catalysts, the merocyanine photoacid benefits from rapid isomerization kinetics and high fatigue resistance.



© PCCL, Principle of reversible activation of the new catalyst (left) and reversible writing of patterns with mask lithography (right).

The exceptionally high level of control achieved over the isomerization state of the photoacid is employed

for the generation of micro-gradients of active catalyzing species. The resulting nonuniform profile of mechanical properties paves the way towards the gradient-driven, mold-free reshaping of free-standing polymer films. Moreover, bending radii are accurately predictable based on a newly established empirical model, designed to be easily transferable to other polymer systems exhibiting nonuniform stress relaxation properties. In addition, the introduced catalytic concept itself can be applied to other acid-catalyzed dynamic polymer networks, rendering them reversibly switchable in terms of a light-controlled ON/OFF behavior.



© PCCL, Local and reversible self-folding of photopolymer films.

### Project coordination (Story)

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Repairecture is a COMET Module Project within the COMET – Competence Centers for Excellent Technologies Programme and funded by BMIMI, BMWET and the provinces Styria and Upper Austria. The COMET Programme is managed by FFG. Further information on COMET: [www.ffg.at/comet](http://www.ffg.at/comet)