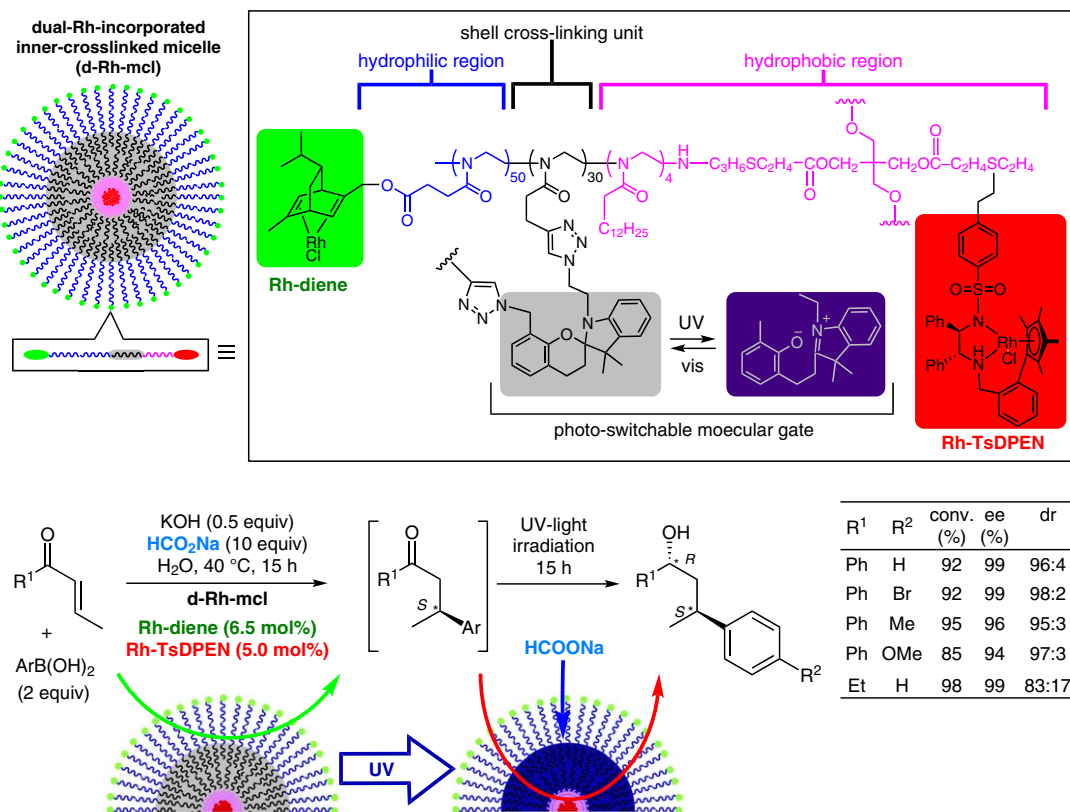


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Compartmentalization and Photoregulating Pathways for Incompatible Tandem Catalysis
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Tandem Asymmetric Michael/Transfer Hydrogenation by Using a Dual Rh-Incorporated Micelle Reactor



Significance: Homochiral Rh–diene and Rh–TsDPEN units were incorporated stepwise into the hydrophilic outer shell and the hydrophobic core regions, respectively, of micelles of a polyoxazoline-based amphiphilic triblock copolymer bearing spiropyran crosslinks (**d-Rh-mcl**). The **d-Rh-mcl** micelles catalyzed an asymmetric 1,4-addition of arylboronic acids to α,β -unsaturated ketones in water (15 h) to give (*S*)-3-arylketone intermediates; successive asymmetric transfer hydrogenation of these (*S*)-intermediates under UV irradiation (15 h) gave the corresponding (*1R,3S*)-3-arylbutanols in high yields and with excellent enantioselectivity.

Comment: The spiropyran cross-linker is a photo-responsive unit that isomerizes to a hydrophilic zwitterionic merocyanine under UV irradiation. Thus, the initial 1,4-addition took place in the hydrophilic corona region of the anchored Rh–diene complex in the absence of UV irradiation. Upon UV irradiation, the zwitterionic merocyanine increased the permeability of the Rh–TsDPEN core region toward water-soluble HCOONa, to promote the transfer hydrogenation in the second reaction step. The 1,4-addition and transfer hydrogenation compete under homogeneous conditions. The photoinduced gating between the compartmentalized catalytic regions was essential for regulating the incompatible multiple catalytic steps.

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