NANOREACTOR

Multiscale modeling of stimuli-responsive nanoreactors

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In Short

- Computer-guided, rational design of smart nanocatalysis
- Molecular simulations of structure and solvation free energies of reactants in polymer networks (hydrogels)
- Coarse-grained Brownian dynamics simulations of reactant transport through hydrogels
- Partial differential equations to describe the diffusion-controlled reaction rate in smart nanoreactors

Abstract: The catalysis by metal nanoparticles is one of the fastest growing areas in nanoscience due to our society's exploding need for fuels, drugs, and environmental remediation. However, the optimal control of catalytic activity and selectivity remains one of the grand challenges in the 21st century. Here, we propose to theoretically derive design rules for the optimization of nanoparticle catalysis by means of thermosensitive yolk-shell carrier systems. In the latter, the nanoparticle is stabilized in solution by an encapsulating, thermosensitive hydrogel shell. The physicochemical properties of this polymeric 'nanogate' react to stimuli in the environment and thus permit the reactant transport and the diffusion-controlled part of the catalytic reaction to be switched and tuned, e.g., by the temperature or the pH. The novel hybrid character of these emerging 'nanoreactors' opens up unprecedented ways for the control of nanocatalysis due to new designable degrees of freedom. The complex mechanisms behind stimuli-responsive nanocatalysis call for a concerted, interdisciplinary modelling approach. In particular, it can only be achieved by combining multiscale computer simulations of solvated polymers with the statistical and continuum mechanics of soft matter structures and dynamics. The key challenge is to integrate the molecular solvation effects and our growing knowledge of hydrogel mechanics and thermodynamics into advanced reaction-diffusion equations for a quantitative rate prediction. Modelling on all scales require parallel computing for molecular dynamics (MD) simulations, Brownian dynamics (BD) simulations, and the optimised solution of systems of partial differential equations. The expected results and design principles will help our collaborators to synthesize tailor-made, superior nanocatalysts and will advance our understanding of their structure-reactivity relationship.

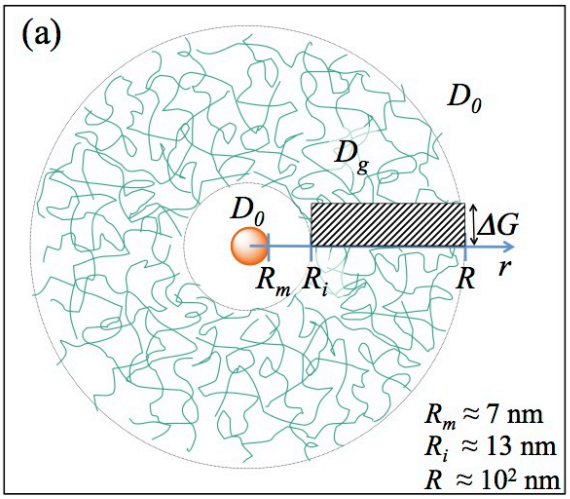

***Figure 1:** Illustration of a yolk-shell nanoreactor: the nanoparticle of radius R_m is encapsulated by a thermosensitive polymer shell with inner and outer radii R_i and R, respectively. The reactants have a diffusion constant D_0 outside and D_g inside the shell, respectively. For penetrating the shell, the reactants require a free energy ΔG. These quantities are key to the understanding and control of nanoreactor catalysis and will be caculated in this project by a multiscale approach, spanning all-atom to coarse-grained simulations and the numerical solution of systems of reaction-diffusion equations.*

WWW

https://www.physik.hu-berlin.de/de/forschung

More Information

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Project Partners

Dr. Yan Lu and Prof. Dr. Matthias Ballauff: experimental partners at the Humboldt-University and the Helmholtz-Zentrum Berlin

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