

Single-Molecule Observation of Diffusion and Catalysis in Nanoporous Solids

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Nanoporous solids, including microporous, mesoporous and hierarchically structured porous materials, are of scientific and technological interest because of their high surface-to-volume ratio and ability to impose shape- and size-selectivity on molecules diffusing through them. Large efforts have been put into the mechanistic understanding of diffusion–reaction relationships of nanoporous solids, such as zeolites, with the ultimate goal of developing materials with improved separation, adsorption, and catalytic performances. Single-molecule fluorescence microscopy can be used to explore the pore space via the trajectories of individual molecules. This ensemble-free perspective directly reveals heterogeneities in diffusion and diffusion-related reactivity of individual molecules, which would have been obscured in bulk measurements. In this lecture, I will discuss the latest developments in the spatial and temporal characterization of nanoporous solids using single-molecule localization microscopy. This will be done by illustrating various aspects of this approach and showcase, using examples from our own research work, how it can be used to follow molecular diffusion and reaction behaviors in nanoporous solids.

Relevant articles from our own research work:

J.J.E. Maris, D. Fu, F. Meirer, and B.M. Weckhuysen, Single-molecule observation of diffusion and catalysis in nanoporous solids, *Adsorption* 2021, **27**, 423.

J.J.E. Maris, F. Rabouw, and B.M. Weckhuysen, F. Meirer, Classification-based motion analysis of single-molecule trajectories using DiffusionLab, *Scientific Reports* 2022, **12**, 9595.

D. Fu, J.J.E. Maris, K. Stanciakova, N. Nikolopoulos, O. van der Heijden, L.D.B. Mandemaker, M.E. Siemans, D. Salas Pastene, L.C. Kapitein, F.T. Rabouw, and B.M. Weckhuysen, Unravelling channel structure-diffusivity relationships in zeolite ZSM-5 at the single-molecule level, *Angew. Chem. Int. Ed.* 2022, **61**, e202114388

F.C. Hendriks, S. Mohammadian, Z. Ristanovic, S. Kalirai, F. Meirer, E.T.C. Vogt, P.C.A. Bruijninx, H.C. Gerritsen, and B.M. Weckhuysen, Integrated transmission electron and single molecule fluorescence microscopy correlates reactivity with ultrastructure in a single catalyst particle, *Angew. Chem. Int. Ed.* 2018, **57**, 257

Z. Ristanović, M.M. Kersters, A.V. Kubarev, F.C. Hendriks, P. Dedecker, J. Hofkens, M.B.J. Roefsaers, and B.M. Weckhuysen, High-Resolution Single-molecule fluorescence imaging of zeolite aggregates within real-life fluid catalytic cracking particles, *Angew. Chem. Int. Ed.* 2015, **54**, 1836.

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Z. Ristanovic, A.V. Kubarev, J. Hofkens, M.B.J. Roefsaers, and B.M. Weckhuysen, Single molecule nano-spectroscopy visualizes proton-transfer processes within a zeolite crystal, *J. Am. Chem. Soc.* 2016, **139**, 13586.

Z. Ristanovic, J.P. Hofmann, G. De Cremer, A.V. Kubarev, M. Rohnke, F. Meirer, J. Hofkens, M.B.J. Roefsaers, and B.M. Weckhuysen, Quantitative 3D fluorescence imaging of single catalytic turnovers reveals spatiotemporal gradients in reactivity of zeolite H-ZSM-5 crystals upon steaming, *J. Am. Chem. Soc.* 2015, **137**, 6559.



