

Mesopore Functionalization as Highly Specific Tool for the Control of Single Molecule Dynamics in Silica Materials

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1. Introduction

Nanoporous channel systems, for example M41S-materials, form a class of very promising host systems in many fields of modern science. Various guests can be embedded into their nanometer-sized pores. Yet, directly influencing the diffusion dynamics of the incorporated guest molecules is not an easy task, even though a broad range of applications in diverse fields could profit from this ability. Organic functionalizations within the porous network of the silica material could be an efficient tool for realizing this aim. Hence, this study examines the effects of such functionalizations onto the diffusion dynamics of a terrylendiimide (TDI) dye on a single molecule level.

2. Method: Single molecule wide-field fluorescence microscopy

The diffusional behaviour of fluorescent molecules is elucidated in our group by sophisticated single molecule microscopy techniques. They have been established within the last decades and have experienced a broad distribution due to their numerous advantages^[1].

In wide-field fluorescence microscopy the whole sample is constantly illuminated. In combination with a state-of-the-art CCD camera images can be recorded with a temporal resolution down to 7ms. This prevails over classical confocal techniques especially for fast diffusing molecules due to avoiding the time-consuming scanning process. The recorded frame sequences provide a unique insight into the trajectories of the single molecules and thereby allow for a direct observation of the underlying molecular mechanisms. In contrast, conventional ensemble techniques would obscure the dynamical behaviour of individual molecules due to the averaging process.

3. Experiment

Within our group substantial knowledge has been gathered about the various aspects of dynamics of dye molecules (translational, orientational, etc.) in porous materials^[2-5].

The work presented here focuses on the investigation of the influence of different organic functionalizations onto the diffusion dynamics of a TDI dye as guest molecule within the mesoporous network of a hexagonal thin silica film (Fig. 1(a)). The films were

synthesized with the non-ionic Block-copolymer Brij 56 as template. Hence, within the pores the template micelles coexist with an organic carpet of the functional groups which are covalently attached to the silica host structure. The interactions of the TDI molecules with the pore inner surface are investigated in detail. For example, the trajectories of diffusing dye molecules can be recorded by tracking their fluorescence spots with high positioning accuracy throughout a sequence of individual widefield images (Fig. 1(b)).

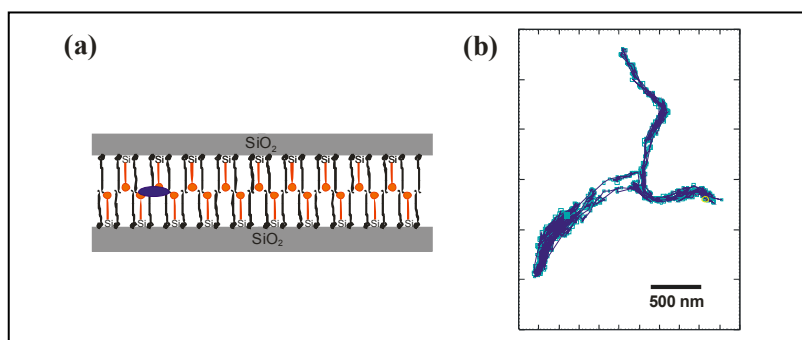


Fig. 1: (a) Diffusion of a TDI molecule within a functionalized mesopore. (b) Single molecule tracking reveals molecular trajectories.

The specific choice of the functional group then allows for a precise tuning of the host-guest interactions and thus the guest dynamics. Furthermore this work will offer detailed mechanistic insights into the diffusion processes within a pore.

The data presented in this work could be of great interest for a variety of applications. Mesoporous silica nanoparticles are discussed in biomedicine as drug-delivery systems. For applications in this field the particles should exert a depot effect, so that the pharmaceutical agent can be dispensed over a preferably long period of time. Hence, the ability of influencing the diffusion dynamics is a crucial prerequisite for the development of such systems. Furthermore, applications in material science as well as catalysis and chromatography could also benefit from the results of this study.

References

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