

Diffusion at tilt grain boundaries in polycrystalline porous materials

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Polycrystalline porous materials are prevalent throughout nature and are also used in many technologies such as catalysis, CO₂ sequestration, gas separation, ceramics and pharmaceutical industry. Inferring the diffusion behaviour of fluids through these materials is critical to understanding the broader phenomena they appear in as well as to fully exploiting their utility. In heterogeneous catalysis, the performance of a catalyst is strongly linked to the diffusivity of guest molecules entering and leaving the catalyst for diffusion limited processes; higher diffusivities generally lead to better catalytic performance of a material. [1] A key aspect of designing heterogeneous catalysts is to reduce diffusion limitations and deactivation within a target catalyst without compromising its material density and, thus, volumetric activity. [2, 3] In CO₂ capture from flue gases and separation technologies, using porous membranes is a much more energy efficient alternative to using conventional methods that often require solvent absorption and desorption, which in turn requires significant heating.

Silicates such as zeolites are a very useful class of materials due to their utility in many of these applications and their microporous structure. There have been many studies of gas diffusion through single crystallites of zeolites using molecular dynamics (MD) simulations. [4, 5] However, a real catalyst particle is usually a composite of zeolite grains, in which sub-micron sized or nano sized crystallites are randomly oriented with respect to each other, thus forming tilt grain boundaries at contact locations. [6] Here we present results of classical MD simulations of diffusion at some model tilt grain boundaries created computationally in α -quartz. Our results show that during the diffusion of CO₂ through polycrystalline silica at high gas pressures, a significant amount of the gas accumulates at the tilt grain boundaries, where it diffuses more slowly than on two-dimensional flat surfaces of the same material. We link these results to the energy profile of the system in an attempt to quantify the extent to which polycrystallinity would affect the performance of a material under various conditions.

References

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