Diffusion of light olefins through ZIF-8 unraveled by machine learning potentials

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In view of the current energy crisis, improvement to highly energy consuming processes is more relevant than ever. One such high energy consuming process is the cryogenic separation of small alkanes and alkenes, which account for around 0.3% of the world's energy consumption.[1] This process involves the cooling of for example ethane and ethene to below their boiling point which is at -89°C for ethane and -103.7°C for ethene. An improvement to this method can be done by introducing a sieving medium, which could lead to a separation of the different substances at room temperature, removing cooling requirements. One such medium, that is investigated, are membranes composed of nanoporous crystalline materials like for example zeolitic imidazolate frameworks (ZIFs). ZIFs are especially interesting based on their high selectivity in separation of ethane from ethene (C2) and propane from propene (C3) species at room temperature.[2,3]

In this work, the impact of ZIF-8 onto the separation behavior of ethane and ethylene is investigated by means of molecular simulations. For this, molecular dynamics (MD) simulations are performed using a state-of-the-art machine learning potential (MLP) derived with Psiflow.[4] The MLP is trained with MD *ab-initio* data gained by use of CP2K.[5] The MLP will be used in combination with the enhanced sampling method called umbrella sampling (US), in which the guest molecule movement is modeled along a collective variable, being the diffusion path in this case. A free energy profile (FEP) can be extracted from the results of the US simulation and relevant properties such as self-diffusion coefficients and free energy barriers can be extracted from the FEP. This will be done for both ethane and ethene separately and a combination of both guest molecules in ZIF-8.

Furthermore, by use of different guest loadings transport- and corrected self-diffusion coefficients can be extracted from the simulations. This advanced insight from both the highly accurate MLP simulations and the extensive analysis of the FEPs can be used to find and/or engineer new host systems with high efficiency in C2/C3 species separation.

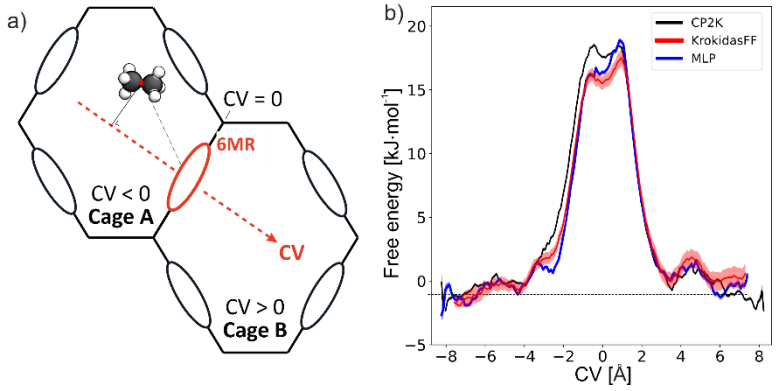

Figure 1: (a) Schematic view of an ethane molecule diffusing in a ZIF-8 host molecule (B) Schematic free energy profile and a sketch of the enthalpic contribution to the free energy profile.

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