Development and application of an advanced percolation model for pore network characterization by physical adsorption

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Physical adsorption is one of the most widely used techniques to characterize porous materials. However, challenges and open questions persist in characterizing disordered and hierarchically structured porous materials. This study introduces a pore network model aiming to enhance the textural characterization of nanoporous materials. The model, based on percolation theory on a finite sized Bethe lattice, includes all mechanisms known to contribute to adsorption hysteresis in mesoporous pore networks. It accounts for delayed and initiated condensation during adsorption as well as equilibrium evaporation, pore blocking and cavitation during desorption. Coupled with dedicated non-local-density functional theory (NLDFT) kernels, the proposed method provides a unified framework for modeling the entire experimental adsorption-desorption isotherm, including desorption hysteresis scans.

The applicability of the method is demonstrated on a selected set of nanoporous silica materials exhibiting distinct types of hysteresis loops (types H1, H2a, H1/H2a and H5), including KIT-6 silica, hybrid SBA-15/MCM-41 silica with plugged pores, a hierarchical meso-macroporous monolith and porous Vycor glass. For all materials, good correlation is found between calculated and experimental primary adsorption and desorption isotherms as well as desorption scans. The model allows to determine key pore network characteristics such as pore connectivity and pore size distributions as well as a parameter indicating the impact of pore network disorder. The versatility and enriched textural insights provided by the proposed novel network model allow for a comprehensive characterization previously inaccessible. It has the potential to provide important guidance for the design and selection of porous materials for optimising various applications, including separation processes, heterogeneous catalysis, gas- and energy storage. [1]

[1] J. Söllner, A.V. Neimark, M. Thommes, Development and application of an advanced percolation model for pore network characterization by physical adsorption, ChemRxiv. (2024) https://doi.org/10.26434/chemrxiv-2024-h9zlm-v2 and under review