VRIJE UNIVERSITEIT BRUSSEL

The faculty of Engineering of the Vrije Universiteit Brussel invites you to attend the public defense leading to the degree of

DOCTOR OF ENGINEERING SCIENCES

of **Niels De Witte**

The public defense will take place on **Monday 26th August 2024 at 4:00 pm** in room **D.2.01** (Building D, VUB Main Campus)

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ADSORPTIVE EFFECTS ON CHA-, AEI-, AND LTA-TYPE SMALL-PORE ZEOLITES

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Abstract of the PhD research

Zeolites are microporous crystalline materials composed of aluminium and silica tetrahedrons that form a network of channels and cavities with regularly sized rings, often referred to as pores. Small-pore zeolites have 8-membered pores, which are the smallest pore size through which hydrocarbon can diffuse. In these small-pore zeolites, the close matching size of the molecules and the zeolite's pore window strongly impacts adsorption, diffusion and catalytic conversion of molecules. This makes small-pore zeolites attractive adsorbents for industrial application such as methanol-to-olefins (MTO) conversion, selective catalytic reduction (SCR) of NOx, or light gas separations.

This doctoral theses explores the adsorptive effects of CHA-, AEI- and LTA-type small-pore zeolites using low- and high-coverage techniques.

First, the sieving properties of pure-silica LTA- and CHA-zeolites were investigated using inverse gas chromatography experiments with hydrocarbons. This study show kinetic effects where the bond angle of the guest molecule plays a key role. Subsequently, breakthrough experiments with small-pore zeolite materials were performed to investigate the potential of small-pore zeolites for light gas separation such as ethane/ethane and propane/propene.

Next, non-monotonic chain-length dependence behaviour of hydrocarbons on AEI and CHA-type zeolites was investigated using n-alkanes and 1 alkenes. These unusual effects, coined cage effects, are mostly computationally studied on all-silica zeolites using n-alkanes as probe molecules. The unusual adsorption have been proposed to originate from the coiling of long molecules in the zeolite's nanocages. Experimentally, they have only been reported for n-alkane adsorption on CHA structures. Such fundamental adsorption effects have been speculated for other, more reactive, chemical groups as alkenes, and for other framework types than CHA, yet have not been reported experimentally. This study is the first to experimentally show cage-effects for 1-alkenes, and highlights the differences with n-alkane adsorption. Furthermore, it is the first study to observe cage-effects for a structure other than CHA, the small-pore AEI structure.

Lastly, the influence of aluminium distribution on the adsorption properties of intermediate- and high-silica CHA zeolites was investigated, as previous studies have shown that the sitting of the aluminium tetrahedrons in the framework affects the catalytic conversion in CHA-type zeolites. The adsorption properties of various samples with differing Al-pairing is reported.

Overall, this PhD research explored and revealed fundamental adsorptive effects in commercially applied small-pore zeolites.