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The Impact of HKUST-1 Metal-Organic Framework Pretreatment on Dynamic Acetaldehyde Adsorption

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Abstract : Volatile Organic Compounds (VOCs) are a real health issue, particularly in domestic indoor environments. Among these VOCs, acetaldehyde is frequently monitored in dwellings 'air, especially due to smoking and spontaneous emissions from the new wall and soil coverings. It is responsible for respiratory complaints and is classified as possibly carcinogenic to humans. Adsorption processes are commonly used to remove VOCs from the air. Metal-Organic Frameworks (MOFs) are a promising type of material for high adsorption performance. These hybrid porous materials composed of metal inorganic clusters and organic ligands are interesting thanks to their high porosity and surface area. The HKUST-1 (also referred to as MOF-199) is a copper-based MOF with the formula $[Cu_3(BTC)_2(H_2O)_3]n$ (BTC = benzene-1,3,5-tricarboxylate) and exhibits unsaturated metal sites that can be attractive sites for adsorption. The objective of this study is to investigate the impact of HKUST-1 pretreatment on acetaldehyde adsorption. Thus, dynamic adsorption experiments were conducted in 1 cm diameter glass column packed with 2 cm MOF bed height. MOF were sieved to 630 µm - 1 mm. The feed gas (Co = 460 ppmv ± 5 ppmv) was obtained by diluting a 1000 ppmv acetaldehyde gas cylinder in air. The gas flow rate was set to 0.7 L/min (to guarantee a suitable linear velocity). Acetaldehyde concentration was monitored online by gas chromatography coupled with a flame ionization detector (GC-FID). Breakthrough curves must allow to understand the interactions between the MOF and the pollutant as well as the impact of the HKUST-1 humidity in the adsorption process. Consequently, different MOF water content conditions were tested, from a dry material with 7 % water content (dark blue color) to water saturated state with approximately 35 % water content (turquoise color). The rough material – without any pretreatment – containing 30 % water serves as a reference. First, conclusions can be drawn from the comparison of the evolution of the ratio of the column outlet concentration (C) on the inlet concentration (Co) as a function of time for different HKUST-1 pretreatments. The shape of the breakthrough curves is significantly different. The saturation of the rough material is slower (20 h to reach saturation) than that of the dried material (2 h). However, the breakthrough time defined for C/Co = 10 % appears earlier in the case of the rough material (0.75 h) compared to the dried HKUST-1 (1.4 h). Another notable difference is the shape of the curve before the breakthrough at 10 %. An abrupt increase of the outlet concentration is observed for the material with the lower humidity in comparison to a smooth increase for the rough material. Thus, the water content plays a significant role on the breakthrough kinetics. This study aims to understand what can explain the shape of the breakthrough curves associated to the pretreatments of HKUST-1 and which mechanisms take place in the adsorption process between the MOF, the pollutant, and the water.

Keywords : acetaldehyde, dynamic adsorption, HKUST-1, pretreatment influence
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