Gas Phase Deposition of ALF-MOF for Selective CO_2 Capture: A Molecular Layer Deposition Study

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The synthesis of alucones, a relatively new class of hybrid inorganic-organic materials, has traditionally been achieved via molecular layer deposition (MLD) using aluminum precursors like trimethylaluminum (TMA) and alcohol-functionalized organic ligands.[1] This method allows for consecutive self limiting gas deposition of organic-inorganic hybrid films with precise control over composition and structure. Alucones are particularly noted for their inherent porosity and low density, making them suitable for applications such as ultra-low-k dielectric films.[2]

Our research presents a novel adaptation of this established process, extending the utility of the alucone deposition technique to the realm of metal-organic frameworks (**MOFs**). Addressing the need for efficient carbon dioxide (**CO_2**) capture technologies, this study presents the pioneering the MLD of aluminum formate **MOF** (**ALF-MOFs**), which is traditionally synthezized via solvothermal methods and is known for its selectively capturing CO_2.[3] Using a commercial ALD tool (Picosun R200), ALF-MOF was deposited from the gas phase through a novel process involving TMA and formic acid (H_2CO_2), **Fig 1a**.

The gowth-per-cycle (GPC) was 3.5 Å for **ALF-MOF**, (**Fig 1b and c)** which is characteristic of an alucone. Mass spectrometric analysis confirmed the polymeric nature of the film, highlighting the repeat unit $Al(OOCH)_3$. Further characterization through Fourier-transform infrared spectroscopy and X-ray photoelectron spectroscopy) elucidated the bridging nature of the FA ligand to the Al nodes.

Quartz crystal microbalance analysis substantiated the step-wise growth of the target **ALF-MOF**. Additionally, the exposure of **ALF-MOF** to a CO_2/N_2 gas stream demonstrated its selective CO_2 capture capabilities. This presentation will delve into the nuances of the deposition and structure of ALF-MOF, emphasizing the nature and reversibility of CO_2 uptake. Our findings represent a significant advancement in MOF deposition, showing a scalable and efficient method for CO_2 capture that could have implications for environmental sustainability and industrial applications.

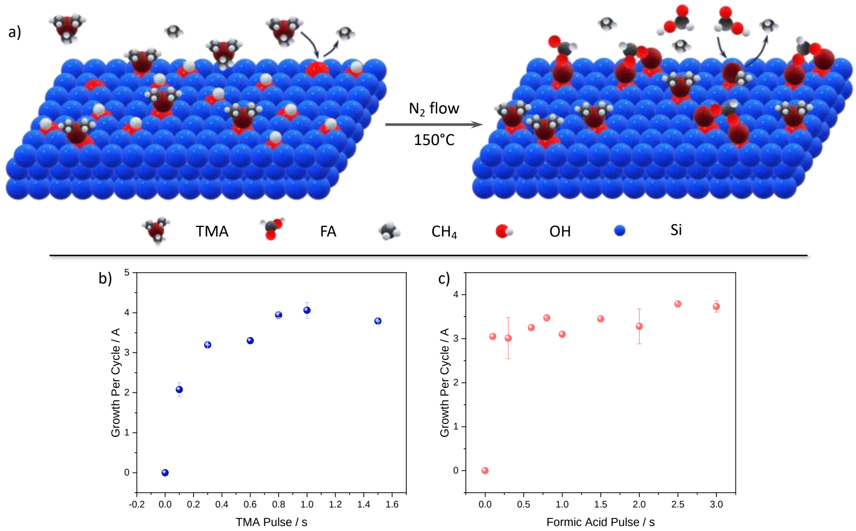

Fig 1. a) Graphical illustartion of AlF MOF deposition, b) GPC of AlF at constant FA pulse and c) GPC of AlF at constant TMA pulse

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