Controlled growth and enhanced processing of 2D Covalent Organic Frameworks for Sustainability Applications

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Covalent organic frameworks (COFs) are highly tunable, crystalline porous materials with significant potential in applications ranging from membrane separations and photocatalysis to ion transport and electrochemical energy conversion. However, their broader adoption is hindered by persistent challenges in synthesis scalability, processability, and integration into functional devices.

This talk outlines recent efforts to leverage the inherent dynamic and reversible chemistry of COFs to address these limitations and advance their practical deployment for sustainable water and energy applications. I begin by introducing a novel synthesis approach that yields stable, homogeneous solution suspensions of crystalline COF particles and colloids, enabling solution-processable formats such as thin films via casting.[1–3] Real-time mechanistic insights into COF nucleation and growth, gained through liquid-cell transmission electron microscopy (TEM) and dynamic light scattering (DLS) studies, reveal how homogeneous conditions influence crystallinity and morphology.[1–3]

Building on these insights, I present a high-throughput continuous-flow synthesis strategy capable of producing highly crystalline COFs in diverse macroscopic forms—including membranes, monoliths, and packed beds—with space-time yields approaching ~60,000 kg m^{-3} day^{-1}.[1,4] This platform overcomes conventional batch-processing limitations and enables direct integration of COFs into scalable systems. The functional potential of these materials is further demonstrated through two case studies: (1) PFOA photocatalytic degradation with approximately 20% conversion to fluoride ions,[1,4] and (2) proton-conductive COF foams functioning as solid electrolytes for electrochemical CO_2 reduction (CO_2RR).[1,5] The latter enables low-voltage CO_2RR operation with selective production of formic acid and C_2+ products at industrially relevant current densities, while maintaining long-term recyclability and stability.

Together, these results establish a path toward COF technologies that are not only chemically precise but also engineerable at scale, enabling real-world deployment in clean water and clean energy systems. This work demonstrates novel processing strategies and applications enabled by COF dynamic chemistry.

References

[1] Khalil, S. Transforming Covalent Organic Framework Synthesis for Advanced Applications: From Solution Processing to High- Throughput Production in Flow Reactors. Rice University, 2023.

[2] Khalil, S.; Meyer, M. D.; Alazmi, A.; Samani, M. H. K.; Huang, P.-C.; Barnes, M.; Marciel, A. B.; Verduzco, R. Enabling Solution Processable COFs through Suppression of Precipitation during Solvothermal Synthesis. ACS Nano 2022, 16 (12), 20964–20974. https://doi.org/10.1021/acsnano.2c08580.

[3] Khalil, S.; Sun, M.; Yang, Z.; Marciel, A. B.; Jones, M. R.; Verduzco, R. In Situ Observation of Covalent Organic Framework Growth in Solution. Small n/a (n/a), e02087. https://doi.org/10.1002/smll.202502087.

[4] Khalil, S.; Alazmi, A.; Gao, G.; Martínez-Jiménez, C.; Saxena, R.; Chen, Y.; Jiang, S.-Y.; Li, J.; Alhashim, S.; Senftle, T. P.; Martí, A. A.; Verduzco, R. Continuous Synthesis and Processing of Covalent Organic Frameworks in a Flow Reactor. ACS Appl. Mater. Interfaces 2024, 16 (41), 55206–55217. https://doi.org/10.1021/acsami.4c09577

[5] Khalil, S.; Kim, J. 'Timothy'; Alazmi, A.; Elgazzar, A.; Gao, G.; Wang, H.; Verduzco, R. Covalent Organic Frameworks as Porous Solid Electrolytes for Electrochemical CO2 Reduction. Advanced Functional Materials n/a (n/a), 2503204. https://doi.org/10.1002/adfm.202503204

Figures

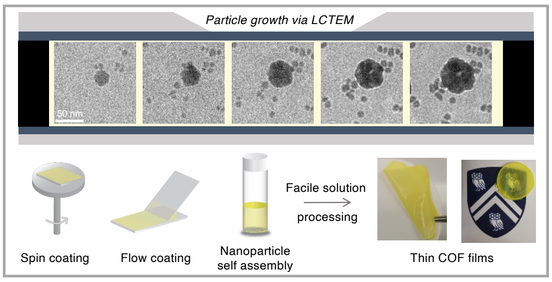

Figure 1: Controlled growth and solution processing of 2D COF nanoparticles.