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Hydrocarbon Frameworks with Long-Range Order

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Introducing long-range order into hydrocarbon covalent organic frameworks (COFs) remains a fundamental challenge because carbon–carbon bond-forming reactions are typically irreversible and lack effective error-correction mechanisms. Here, we demonstrate a molecular approach to enhance the crystallinity of a hydrocarbon framework.[1] A fully hydrocarbon two-dimensional (2D) COF, HKU-50, was synthesized via reversible olefin metathesis. Polymerization of a vinylene-bearing monomer using the second-generation Grubbs catalyst initially yielded an amorphous network. Upon addition of trans-stilbene, secondary metathesis is activated, regenerating catalysts that are otherwise trapped within the growing polymer. This enables continuous bond exchange and error correction during framework formation. As a result, the system evolves toward its thermodynamically favored product, HKU-50, with long-range order. The resulting COF exhibits high thermal and chemical stability, along with enhanced photophysical properties, including red-shifted emission, prolonged fluorescence lifetime, and a 4-fold increase in photoluminescence quantum yield relative to its amorphous analogue due to its extended order. These findings demonstrate a molecular approach that actively controls the crystallinity of hydrocarbon frameworks without the use of supports or interfaces that induce partial ordering of the building units.

References:

[1] Sui, X.; Xiong, C.; He, Jiaxing; Park, S.; Kong, F.;Ying Z.; Jiang, E.; Chow, P.; Zhou, Y.; Jeong K.; Terasaki, O.; Han, Yu.; Jeong, N. *; Phillips, D. *; Lee, S.* *J. Am. Chem. Soc*. **2026**, Just Accepted. https://doi.org/10.1021/jacs.5c22787.