Abstract: Synthetic permanently porous materials are poised to play a key role in our transition to a more sustainable society. Owing to their structural and chemical modularity, synthetic frameworks, such as metal–organic, covalent, and hydrogen-bonded networks, are uniquely amenable to realizing highly specific functionality for emergent clean energy applications. However, desired performance for many pressing challenges, such as H2 delivery and light alkene purification, continue to elude established framework classes. Thus, the pursuit of synthetic porous frameworks with emergent behaviors and of entirely new classes of frameworks is essential. In this vein, this talk will cover two distinct efforts to diversify function and form. First, recent efforts to harness reversible framework flexibility to enhance usable capacities of hydrogen storage and delivery and for direct purification of propylene will be discussed. Then, the recent discovery of a novel class of frameworks assembled and stabilized through noncovalent chalcogen bonds, called Chalcogen-Bonded Organic Frameworks (ChOFs), will be detailed.