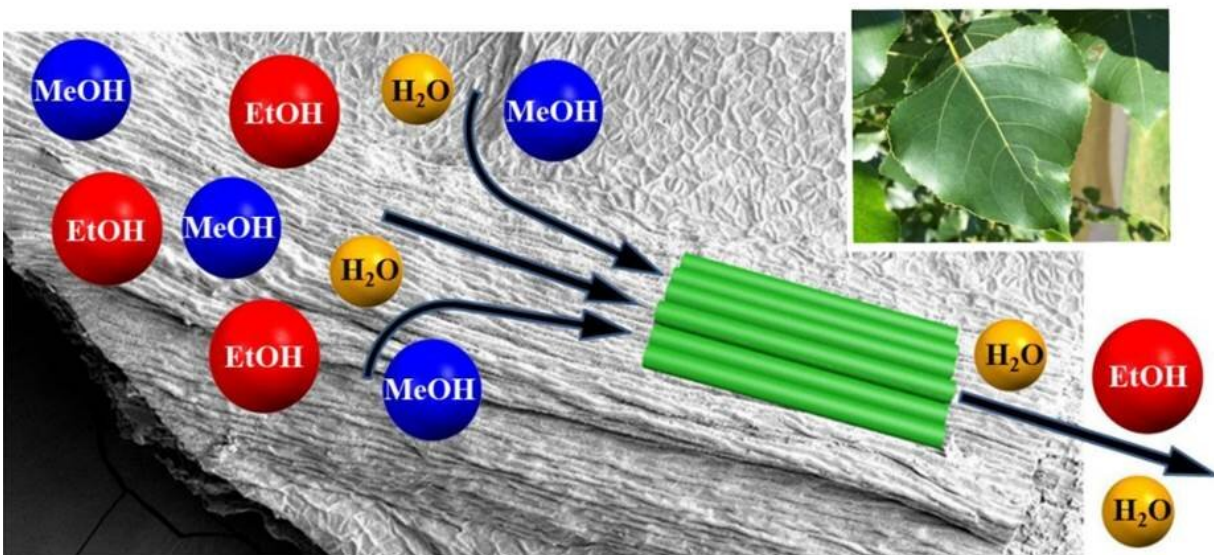


Enhancing the performance of metal-organic framework materials

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Micrograph photo (total width approximately 1,5 mm) showing the novel adsorbent material that was produced using a poplar leaf as a template. The channel architecture is clearly noticeable. The drawings show the tubular structure (green) and the molecular separation process. Credit: HIMS

Researchers at the group of Dr. Stefania Grecea at the University of Amsterdam's Research Priority Sustainable Chemistry have devised a way to enhance the practical performance of metal-organic frameworks (MOFs). By using leaves from the black poplar as a template, they produced hierarchical porous structures of mixed-metal oxide materials

that can act as support for MOF crystals. In a recent edition of the journal *ACS Applied Materials & Interfaces*, Ph.D. student Yiwen Tang, in collaboration with Dr. David Dubbeldam of the UvA Computational Chemistry group, demonstrate the unique adsorption and separation properties of the bio-inspired design.

Separation of water-alcohol mixtures is one of the most challenging problems associated with the practical application of bioethanol as a sustainable fuel. Produced from agricultural feedstocks, algae farms or the fermentation of molasses, bioethanol contains both water and methanol as impurities. Obtaining fuel-grade bioethanol from these water-alcohol mixtures using traditional distillation is not practical because water and ethanol form a so-called azeotropic mixture.

The cost-effective and green alternative to distillation is adsorptive separation. In biofuels production, this method relies on the development of adsorbent [materials](#) which are highly selective towards ethanol or the impurities in the mixture. At the University of Amsterdam's Research Priority Area Sustainable Chemistry, the group of Dr. Stefania Grecea develops synthetic approaches for designing porous molecular-based materials with such selective adsorption properties.

Adsorbent Materials

Suitable adsorbent materials for separation applications should have an appropriate porous structure and high specific surface area to facilitate both the adsorption and diffusion of specific molecules. A specific class of adsorbent materials are metal-organic frameworks (MOFs). They have a high specific area and by tuning the size and functionality of their pores at the [molecular level](#), specific adsorption selectivities can be achieved.

However, practical application also depends on their macroscopic

properties. Often MOFs are synthesized as powders of tiny crystals. These cannot be used directly in industrial applications because they have limited packing density as well as high diffusion barriers. A solution is to shape MOFs as granules, pellets or monoliths, or to disperse them within thin films, creating membranes. However, the pressure applied in such shaping methods leads to loss of crystallinity and therefore to reduced activity or even inactivation of the MOF materials. It is therefore still quite a challenge to find the most appropriate processing method of MOFs.

Looking at leaves

In search for ways to improve MOF performance, the Amsterdam researchers turned to nature, in particular, to green plant leaves. Scientists have already used natural leaves as templates to design heterogeneous photocatalysts, as they are structured to provide efficient light harvesting. Such artificial leaf structures have proven to be very effective for hydrogen production.

The UvA researchers took their inspiration from the natural leaf vein system that has evolved for transporting aqueous liquids. It is a hierarchical porous system consisting of many fibres and vessels of different sizes. In separation technology, hierarchically porous materials having multi-level pores often present enhanced adsorption performance compared to uniformly-sized porous materials.

Therefore, the researchers synthesized a mixed-metal oxide material with a hierarchical porous structure using a sol-gel method in which natural leaves of the black poplar (*Populus nigra*) were used as a template. This mixed-oxide artificial leaf was then used as a support for creating a homogeneously dispersed layer of MOF crystals.

Detailed morphological studies showed that the resulting composite

material indeed has a hierarchical porous structure and that MOF crystals with narrow size distribution are homogeneously dispersed at the inner surface of the hierarchical pores.

Performance studies

In the light of application in bioethanol purification, Yiwen Tang studied the water, methanol and ethanol adsorption properties of the new material. He established that the selectivity varies in the order methanol > ethanol > water. Subsequent molecular simulations performed by Dr. David Dubbeldam using equimolar ethanol-methanol mixtures showed that methanol adsorption is highly selective in the low-pressure range. Moreover, the material is effective in separating water-ethanol mixtures with ethanol being adsorbed selectively in the low-pressure range, whilst water is adsorbed selectively at high pressures.

The researchers conclude that their bio-inspired synthetic approach is highly relevant not only for molecular separations applications but also as a general strategy for designing MOF composite materials for various applications, including catalysis and molecular sensing.

More information: Yiwen Tang et al. Efficient Separation of Ethanol–Methanol and Ethanol–Water Mixtures Using ZIF-8 Supported on a Hierarchical Porous Mixed-Oxide Substrate, *ACS Applied Materials & Interfaces* (2019). [DOI: 10.1021/acsami.9b02325](https://doi.org/10.1021/acsami.9b02325)

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