Design of novel MOF/palladium composite membranes for hydrogen separation

M. Drobek*, S. Sayegh, L. Badouric, J. Cartier, C. Charmette, A. Julbe and M. Bechelany
Institut Européen des Membranes, CNRS, University of Montpellier, France
*martin.drobek@umontpellier.fr

Considering a significant importance of hydrogen (H_2) in the development of upcoming transportation technologies and numerous chemical processes, there is a pressing need to advance the progress of reliable, swift, and efficient approaches for its optimal separation/purification.

To address this subject, many membrane materials have been considered for selective H_2 extraction, such as purely inorganic or polymeric membranes and their composite or hybrid analogues. For the latter, huge efforts have recently been made to design supported Metal Organic Frameworks (MOFs), suitable for either gas adsorbers with high accessible surface or selective membrane barriers. Among different families of MOFs, special attention has been paid to ZIF (Zeolitic Imidazolate Framework) structures, with particular focus on ZIF-8 having a huge potential as a molecular sieve for gases. As for inorganic membrane materials, in addition to metal-oxides, strong emphasis has been placed on metal-based membranes, such as palladium (Pd) and its alloys which feature particularly attractive selectivities enabling the production of pure H_2.

In order to merge the unique properties of MOFs as efficient microporous molecular sieves and those of Pd as a highly selective material towards H_2, herein, we take advantage of our expertise to propose an original synthesis strategy based on the solvothermale conversion of metal oxides and Atomic Layer Deposition (ALD) of both *i)* ultrathin oxide layers with controlled thickness and *ii)* Pd nanoparticles with controlled size. The former serves as a sole source of metal that is subsequently converted to MOF during the solvothermal synthesis step, while simultaneously confining the Pd nanoparticles within the resulting MOF membranes.
Thus, this synthesis strategy enables the fabrication of H_2 selective MOF/Pd composite membranes presenting a double level of confinement with *i)* Pd nanoparticles dispersed homogenously in the ZIF-8 matrix and *ii)* the ZIF-8/Pd composite material fully confined in the macropores of the ceramic tubular support. The resulting composite membrane material features attractive separation properties for the extraction of H_2 from gas mixtures containing namely N_2 or CO_2 with promising selectivities.

The results that will be presented demonstrate the proof-of-concept for the design of novel H_2 selective composite membranes. Furthermore, the developed synthesis strategy could be extended to the preparation of other three-dimensional membrane architectures by a judicious choice of metals and appropriate MOF structures, thus opening new avenues for the preparation of highly selective separation/catalytic devices.

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