12th International Conference on Inorganic Membranes,
University of Twente, Enschede, Netherlands,
July 9-13, 2012

Hydrogen separation from the gas mixtures with composite membranes based on Group 5 metals

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The chemical energy of fossil fuels can be directly converted into electricity with using hydrogen fuel cells (e.g. PEMFC). For this purpose the fossil fuels are converted into a hydrogen-rich gas mixture from which hydrogen with the purity of no less than 99.999% should be extracted for feeding PEMFC. Typically, Pd alloy membranes are used for that. However the price of such membrane system is extremely high. The membranes of Group 5 metals covered with Pd (e.g. Pd-V-Pd or Pd-Nb-Pd) could extract hydrogen with high rate and infinite selectivity. The extremely fast hydrogen transport through Group 5 metals is caused by both the very high hydrogen diffusivity and the great hydrogen solubility. The latter, however, leads to a number of problems when these metals are used as membrane materials:

(1) Hydrogen concentration reaches critical limit already at H2 pressure 5–10 bar at $T \leq 400°$ C where the Pd coating remains to be thermally stable. That limits the application field of the composite membranes, e.g. it is problematic to produce hydrogen at high enough pressure required for operation of alkaline fuel cell;

(2) The great hydrogen dilatation caused by high hydrogen concentration may result in failure of tightness of connections between the membranes and the structural parts;

(3) The hydrogen dilatation may lead to a destructive mechanical stress caused by hydrogen concentration gradient across the membrane;

(4) One should take special measures for hydrogen removal from the membrane system at its cooling to prevent the membrane damage due to hydride formation.

In our experiment hydrogen permeability of disk shaped niobium membrane covered with thin Pd layer with magnetron sputtering technique (Pd-Nb-Pd) was one order of magnitude higher than that of palladium membrane of the same thickness. Vanadium based membrane of tubular shape covered with palladium from both sides (Pd-V-Pd) with electroless plating also demonstrated very high hydrogen permeability. This membrane was successfully employed for hydrogen separation from syngas. Hydrogen cycling test (100 cycles, 6 bar) demonstrated high mechanical stability of the membrane in hydrogen atmosphere. Thermal stability test showed that noticeable membrane degradation starts at 500° C because of V interdiffusion to Pd. The module containing 17 tubular Pd-V-Pd membranes was assembled and tested. Thus the composite Pd-V-Pd membranes demonstrated their capabilities of successful operation in spite of the problems mentioned above.

Still the hydrogen solubility in group 5 metals would be desirable to optimize by alloying with other metals. The problem is that the alloys of group 5 metals are usually not ductile enough to produce foils or tubes from them. The authors hope to solve the problem and then they plan to produce membranes of planar and tubular shape of the alloys with optimized hydrogen solubility.

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