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PII: S0376-7388(18)31099-8
DOI: <https://doi.org/10.1016/j.memsci.2018.06.008>
Reference: MEMSCI16225

To appear in: *Journal of Membrane Science*

Received date: 24 April 2018
Revised date: 26 May 2018
Accepted date: 5 June 2018

Cite this article as: T.A. Peters, P.A. Carvalho, J.F. van Wees, J.P. Overbeek, E. Sagvolden, F.P.F. van Berkel, O.M. Løvvik and R. Bredesen, Leakage evolution and atomic-scale changes in Pd-based membranes induced by long-term hydrogen permeation, *Journal of Membrane Science*, <https://doi.org/10.1016/j.memsci.2018.06.008>

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Leakage evolution and atomic-scale changes in Pd-based membranes induced by long-term hydrogen permeation

T.A. Peters¹, P.A. Carvalho¹, J.F. van Wees², J.P. Overbeek², E. Sagvolden¹, F.P.F. van Berkel², O.M. Løvvik¹, R. Bredesen¹

¹ SINTEF Industry, P.O. Box 124 Blindern, N-0314, Oslo, Norway

² Energy Research Centre of the Netherlands (ECN part of TNO), P.O. Box 1, 1755 ZG Petten, The Netherlands

Abstract

The long-term operation of a ceramic-supported 5 micron-thick Pd membrane at 450 °C and 27 bar with a feed representing steam-methane-reforming conditions has been monitored over a period of 100 days. Subsequently, a thorough characterisation of the exposed membrane has been performed to investigate leakage behaviour and microstructural changes to the membrane occurring during the long-term membrane operation. Initially, a H₂ permeance of $2.8 \cdot 10^{-4} \text{ mol} \cdot \text{m}^{-2} \cdot \text{s}^{-1} \cdot \text{Pa}^{-0.5}$ and a H₂ permeate purity of 99.8% have been obtained. During continuous operation over 100 days, however, a slow but gradual decrease in H₂ purity has been observed at a close-to-constant H₂ permeance. Post-process leakage behaviour has indicated that the increase in leakage is mainly caused by an increase in the Knudsen flow contribution, reflecting an increase in either the amount or size of nano-sized defects. A rising water test shows that these nano-sized defects are almost evenly distributed over the membrane length. Observation of the membrane cross-section revealed the formation of cavities that grew to sizes up to tens of nanometers, as a result of molecular recombination of hydrogen at low-energy sites such as dislocation walls and high-angle grain boundaries. The spherical shapes of the bubbles point to these being energetically favorable, which may be consistent with comparably high surface tensions. Therefore, the cavities observed are believed to be filled with hydrogen and stabilized by an internal pressure, that effectively counteracts the strong effect of surface energy at these small scales. The cavities may represent the origin of pinhole formation, and the atomic mechanisms behind the observed behavior are discussed and directions for enhancing the microstructural stability of Pd-based membranes are pointed out.

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