**ON THE EFFECT OF STEAM AS SWEEP GAS IN PALLADIUM SUPPORTED MEMBRANE**

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**Highlights**

* Steam induces additional mass transfer limitation in the alumina porous support
* Hydrogen permeation is strongly influenced by steam in the permeate side
* High temperatures reduce the mass transfer resistance induced by steam

**1. Introduction**

In recent years palladium membranes received increasing interest due to their extremely high selectivity and permeability and for the possibility to integrate into membrane reactors for hydrogen purification and production. There are two possible ways to increase the permeation of hydrogen at fixed feed pressure: decreasing the thickness of the membrane to increase the permeance, at the expenses of the selectivities, or to raise the driving force of the process via either a vacuum or a sweep gas in the permeate side of the membrane. Steam is generally suggested as sweep gas as it can afterward be easily separated by condensation. The effect of steam on the permeate side of a membrane is however not fully understood and it is the topic of this work.

**2. Methods**

An ultra-thin Pd-Ag membrane was deposited on asymmetric porous α-alumina tubes (100 nm pore size, from Rauschert) using simultaneous electroless plating (ELP) technique [1]. Thicknesses were defined by controlling the time of plating process. The membranes have been sealed and placed in the setup for its permeation characterization. The gases have been fed in the membrane module through mass flow controllers that can work until 10 l/min. The permeate side of the membrane has been analysed with flow meters in order to determine the gas permeances (H2, CH4, H2O, N2) and with gas chromatographer for measuring the H2 purity in the permeate side. After a pure gas test to calculate the permeance and the selectivity of the membrane, a mixture of 10% H2-90% CH4 was fed in the retentate side while in the permeate side, counter-current sweep gas was applied. A validated model for the description of the membrane, which includes concentration polarization in the retentate, permeate and mass transfer limitation in the porous support was used for comparing and understanding the experimental results [2]. N2 and H2O was applied in the permeate side changing the sweep gas flow rate and the steam concentration from 0 to 93.5%.

The working temperature was changed from 350 °C to 500 °C and the retentate pressure between 1 and 5 bar.

**3. Results and discussion**

As depicted in Figure 1, the negative influence of steam as sweep gas is deduced because of the decreasing hydrogen permeation obtained when steam flow rate is increased. An additional effect apart from the concentration polarization and the mass transfer limitation in the porous support is clearly present in case of steam. Indeed, as shown in Figure 2, the model (that included a dusty gas model for the permeate side) could predict the behavior of the membrane in case of nitrogen as sweep gas but overestimates the hydrogen permeation in the presence of steam as sweep gas. The possible explanation could be found in the adsorption capacity of alumina which it could be relevant even at 400 °C. The XRD of the alumina support sample, moreover shows Si traces, which could represent an additional reason for the adsorption capacity of the support. Therefore, the temperature plays an important role because the lower the temperature, the more remarkable the negative steam effect. At 500 °C the steam adsorption on the alumina support seems to be negligible and indeed the model then predicts well the experimental results.

 

**Figure 1.** Hydrogen permeation trend with steam concentration

**Figure 2.** Comparison between experimental and model results in presence of N2 and H2O as sweep

**4. Conclusions**

The driving force of a Pd membrane when feeding steam as sweep gas is decreased between 350-450 °C because of steam adsorption on the support. Indeed the temperature plays an important role on the decreased hydrogen permeation in presence of steam. No influence of the steam on the retentate side was found in the experimental results.

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