Nanoporous silica membranes with high hydrothermal stability

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Towards hydrothermally stable silica membranes

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Background

Silica membranes for hydrogen separation are asymmetric systems consisting of a macroporous support, an intermediate layer and a thin gas-selective top layer. The structure of a silica membrane is shown in Figure 1.

This type of membrane allow to separate the small hydrogen molecules from larger molecular species, as CO2 and CH4 (Figure 2). Therefore these devices appear to be promising separation systems for the upcoming technology platforms for green fuel production.

However, several works report for this membranes poor stability in presence of steam at temperature as low as 60 °C. As shown in Figure 3, during hydrothermal exposure, the porous silica structure collapse, yielding a denser material with a consequent loss in membrane permeability and selectivity.

Objective

Hydrothermal stability of silica membranes can be effectively improved by doping with various transition metal ions. However the impact of dopant type and concentration on the structure, stability and permeability of silica-based membranes is not clear yet, and the development of these membranes is mainly attained by an empirical approach.

In this study, surfactant micelles were used as templates to tailor 1-2 nm pores in sol-gel derived silica-based materials. Despite such pores are larger than those of hydrogen-selective membranes, this approach allowed comparing materials with different composition, but similar pore structure. Doped and undoped powders were characterized before and after hydrothermal exposure.

Results

After hydrothermal exposure, the unsupported membranes presented lower pore volume, lower surface area and broader pore size distribution. This structural modification was more pronounced for the pure silica membranes than for the doped materials (Figure 4).

The gel-to-glass transition (Tg) temperature were determined by calorimetric analysis (Figure 5).

This work provide bases for the effective design of highly stable silica membranes.

Conclusions

As shown in Figure 6, a good correlation was found between the glass transition temperature of these materials and their surface area loss due to steam-exposure. These results indicate that dopants acted as network formers. Thus, the higher glass transition temperature and the enhanced hydrothermally stability of doped sample can be considered as a result of the higher network connectivity.

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