

Effects of Water Gas Shift Gases on Pd-Cu Alloy Membrane Surface Morphology and Separation Properties

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Abstract: Palladium-copper alloy membranes (2-4 μm thick), which were deposited on porous ceramic tubular supports by electroless plating, separated H_2 from a water gas shift (WGS) mixture at 623-723 K. However, within 120 min at 623 K, the total permeance increased and membrane selectivity decreased. H_2 , N_2 , or He did not change the transport properties of the membrane before WGS gas exposure. Annealing in CO_2 and CO at 523-723 K increased the height of micronscale conical hillocks and defect sizes on the membrane surface by at least a factor of 3. Annealing in H_2 and He after CO and CO_2 exposure decreased the hillock heights by half. The hillocks were clusters of grains with defined valleys. The membrane defects allowed gases other than H_2 to permeate through the membrane. Surface topology changes are partially due to the removal of C impurities by CO_2 to form CO. Hillock heights on 25- μm -thick cast and rolled Pd-Cu alloy foils, which had no C impurities, increased by a factor of 4 at 723 K in the presence of CO_2 . The surface of the electroless-plated films was a factor of 3 rougher than the foils. Decreasing the membrane surface roughness, increasing the membrane thickness, and minimizing C impurities decreased membrane defect formation associated with surface rearrangement. Fewer vacancies and lattice defects in the alloy lattice may also make the foil more resistant to atom rearrangement than the electroless-plated membranes. The extent of WGS, CO disproportionation, and methanation reactions on the membrane increased at higher Cu alloy concentrations. Exposure to CO and CO_2 segregated Pd to the feed side of the membrane and changed the membrane alloy composition and phase structure. The change in phase structure from bodycentered cubic to face-centered cubic decreased the H_2 permeance through the membrane and may increase surface rearrangement.

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