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MS-3-P-2826 TEM characterization of H_2-seclective Pd-based membranes

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Palladium (Pd) and Pd-based membranes have been widely studied for several decades due to their unique hydrogen separation capabilities, and have a large market potential for production of high-purity H_2 fuel for application in e.g. power generation with CO_2 capture, fuel cells or chemical industry. The most common membrane compositions are pure Pd, and alloys of Pd-Ag, Pd-Cu and Pd-Au, where the alloying element first of all increases H_2 permeability or reduces the H_2 flux inhibition due to CO and H_2S.

An understanding of the impact of segregation and grain growth is crucial to design Pd-alloys for various applications. In the current study, the effects of selected treatments, like H_2 permeation and H_2S exposure, on free-standing Pd-alloy samples are characterized using JEOL 2100F operated at 200kV. The SINTEF-patented two-step technique of magnetron sputtering was used to prepare Pd-alloy samples. In this study, the $Pd_{81}Cu_{18}Ag_1$ alloy, relevant for the development for highly permeable sulfur-tolerant Pd-Cu alloy membranes, has been used as model system.

Depending on the information sought, the samples can be studied in TEM in plan-view or in cross-section geometry. Plan-view observation is mainly used to study surface morphology, crystal orientation and chemistry. Compared with the plan-view, cross-section observation provides more information from real-space structures, including defects and interfaces with resolution down to atomic scale.

TEM characterization shows that the substrate-interface of the membrane is smoother than growth-interface. In the as-grown membrane, the grains at or near the substrate-interface are tiny and start to be orientated up at about 50nm from the surface, most grains are distinctly columnar after a growth thickness of about 100nm. The grains at the growth-interface are columnar and become much larger with 50nm in average size. The grain structure of the sputtered films thus reflects the growth process. Under the sputtering conditions applied, the nucleation density on the Si wafer substrate is high, and the smooth nature of the wafer surface also affects the resulting roughness on the substrate side. As more material is deposited, some grains grow while others are terminated and covered. The result is a grain size gradient throughout the film, with elongated grains of preferential orientation along the [111] direction. After a H_2 permeation test the grains grew at both the substrate-interface and the growth-interface to about 100nm. The growth at the substrate interface was most significant. Some voids less than 10nm along the grain boundaries have also been observed. These voids do, however, not contribute to unselective leakage flux, but still, the long-term effect on stability requires further investigation.

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