Preparation of $Pd/TiO_2/Al_2O_3$ composite membranes with high H_2-permeability

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With the rapid development and worldwide utilization of hydrogen energy, Pd composite membranes (including Pd-based alloy ones) have attracted more and more attentions and been regarded as one of the most practical function materials for hydrogen separation in many integrated energy systems [1,2]. To further accelerate the application in the fields of hydrogen separation and purification, palladium composite membranes with high permeability towards hydrogen are extremely demanded; meanwhile, ideal hydrogen selectivity is required as well [3,4].

In our present work, macroporous Al_2O_3 tubes were employed as membrane substrate due to their wide market sources, stable chemical properties and low cost. A novel method for substrate surface modification was introduced for the preparation of highly H_2-permeable palladium composite membranes. Homogeneous suspension comprised of TiO_2 nano-particles and polyvinyl alcohol (PVA) was used for dip-coating treatment on the substrate surface. Afterward, the coated substrate was heated at 600 °C under the atmosphere of nitrogen, resulting in a carbon-doped TiO_2 modified layer on the substrate (i.e., TiO_2–C/Al_2O_3) [5].

For the deposition of palladium membranes on the surface of TiO_2–C/Al_2O_3, electroless plating was performed at room temperature. After completing the deposition, the carbon residue was finally burned out, resulting in a Pd composite membrane denoted as $Pd/TiO_2/Al_2O_3$–1. For comparing, TiO_2/Al_2O_3 was achieved through the conventional solid sintering process in air, and the other Pd composite membrane (i.e., $Pd/TiO_2/Al_2O_3$–2) was prepared. Membrane performance was evaluated by H_2/N_2 single-gas testing at the temperature in range of 350–450 °C.

Experimental results indicated that hydrogen permeability of $Pd/TiO_2/Al_2O_3$–1 was much higher than that of $Pd/TiO_2/Al_2O_3$–2 under the same condition, which could attribute to the increase of effective membrane area in permeate side after the elimination of carbon residue. Moreover, $Pd/TiO_2/Al_2O_3$–1 had a higher selectivity towards hydrogen, indicating that the novel method for membrane preparation significantly facilitated to the decrease of membrane defects, particularly the role of carbon-doped intermediate layer. Long-term testing indicated that the prepared Pd composite membrane was permeable and stable.

Scanning electron microscopy (SEM) images of the surface and cross-section of $Pd/TiO_2/Al_2O_3$–1 were shown as following. It can be seen that there was no visible pinholes in membrane surface, and average membrane thickness was measured to be 4~5 μm.

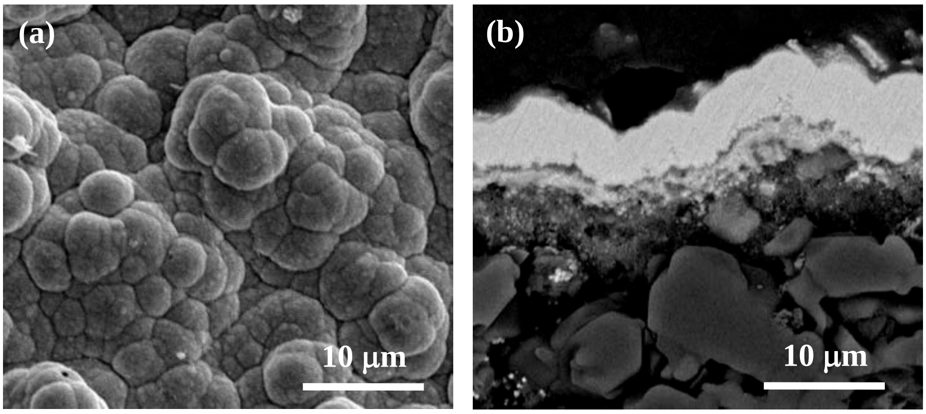

Figure (a) and (b): surface and cross-section SEM images of $Pd/TiO_2/Al_2O_3$–1

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