S-P14

A study of reliability in Pd-Cu-Ni ternary alloy membrane for hydrogen separation

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A Pd-Cu-Ni ternary alloy membrane was fabricated by sputtering and a Cu reflow process. The sputtering method has been used for deposition of a thin, impurity-free Pd alloy membrane. However, the low selectivity of hydrogen has been a major drawback for commercialization of membranes prepared by sputtering methods. In this work, a Cu reflow process was introduced as a solution to this problem. The adoption of the Cu reflow process makes possible a dense and void-free surface, and leads to a dramatic increase of the selectivity of hydrogen to an infinite level. The crystal structure and surface micro-structure of the membrane were subjected to high temperature X-ray diffraction and field emission scanning electron microscope analyses to investigate the effect of Cu reflow on the Pd-Cu-Ni ternary alloying process. The nickel element of the support actively diffused into the Pd-Cu film layer. As a consequence, the Pd-Cu film and Ni support layer were completely integrated as an alloy membrane with a functional gradient. The results of a permeation test using a single gas (H_2 and N_2) at a pressure difference of 280kPa indicated that the selectivity of hydrogen was at an infinite level due to the void free and dense surface of the membrane. The permeability was revealed to be 0.077 $mol \cdot m^{-2} \cdot s^{-1}$. The performance of the membrane formed by sputtering and the Cu reflow process was not degraded after a long working-period of 10 days (250hours) at severe conditions of various temperature and pressure difference. This is most likely due to the excellent stability of the Pd-Cu-Ni ternary alloy, as it is composed of metals with high mutual chemical affinity and is has been fabricated under dry vacuum conditions without any contamination by impurities.