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Degradation mechanism of a Pd/Ta composite membrane: Catalytic surface fouling with inter-diffusion

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ABSTRACT

Composite metallic membranes comprising Pd coated on group 5 elements such as V, Nb, and Ta are actively explored as a promising material for hydrogen permeable membrane. However, such composite membranes are known to suffer from performance degradations owing to excessive surface fouling due to inter-diffusion between Pd and group 5 elements at high temperatures. In this study, mechanically stable Pd/Ta composite membranes are fabricated, and their H_2 permeation flux degradation rates and mechanism of membrane degradation are investigated. Fabricated membranes had full coverage of Pd on Ta surface, with less than 5 wt% of impurities. H_2 permeation rates of Pd/Ta membrane samples with different Pd and Ta thicknesses are evaluated at temperatures from 450 °C to 650 °C for a prolonged period of 20–100 h. Over the tested period, degradation in H_2 permeation rate was 5% or higher, with temperature being a dominant factor. SEM, TEM, and XRD analyses of the surface and cross-sectional morphology, compositions, and crystallinity reveal that the main degradation mechanism of the composite membrane is the inter-diffusion between Pd and Ta leading to the formation of intermetallic compounds as well as morphology change of the catalytic layer. By evaluating the exponential factor in Sievert's equation over the degradation period, it was found that the diffusion mechanism does not change over time, confirming that the bulk diffusion mechanism is affected by the inter-diffusion. H_2 flux degradation was less prominent at lower temperatures with thicker Pd catalytic layers.

1. Introduction

Pure H_2 with high purity (>99.99%) has many applications such as ammonia synthesis, methanol production, fuel processing, energy carrier, etc. Currently, most of H_2 is produced from thermochemical reforming processes such as steam methane reforming. Conventional reforming processes produce not only H_2 but also byproduct gases such as CO_2, CO, and H_2O, necessitating hydrogen purification processes. Of note, H_2 has been emerging as a clean renewable energy carrier with the adoption of more carbon-neutral electrochemical production methods such as electrolysis [1]. However, due to temporal and spatial discrepancies between H_2 production and utilization sites, it is proposed to store H_2 in a form of chemical hydrides [2] such as methanol [3], ammonia [4,5], formic acid [6], and liquid organic hydrogen carrier [7]. To extract H_2 again from the chemical hydrides, reforming processes are required which produce not only H_2 but also byproduct gases, alike conventional reforming processes. Therefore, H_2 purification is an essential process that has the potential to determine the economic viability of H_2 for both chemical feedstock applications in a near term and energy applications in the long term.

To separate H_2 from gas mixtures, pressure swing adsorption

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