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Unconventional hydrogen permeation behavior of Pd/BCC composite membranes and significance of surface reaction kinetics

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ABSTRACT

As one of hydrogen purification techniques, membrane separation has a significant potential to directly obtain high-purity hydrogen from the mixed gas produced from various catalytic reactions. Specifically, dense metallic membranes exhibit several advantages including good mechanical strength under pressurized conditions, thermal stability, and high hydrogen selectivity. Conventionally, their permeation behavior is predicted from a numerical model known as the Sievert's law, which describes diffusion of hydrogen atoms through a metal layer. This study questions the validity of previous permeability trends of the Pd/BCC composite membranes and reveals the importance of surface reactions that significantly affect the permeation behavior of such membranes. A new permeation model developed, considering both the surface reactions at the catalytic layers and bulk diffusion through the metal layers, exhibits good correlation with the experimental permeation characteristics of the Pd/BCC composite membranes. Moreover, the diffusivity coefficients of BCC metals as a function of temperature is determined with higher accuracy than those reported in previous studies having temperature range inconsistency between hydrogen solubility and diffusivity. The experimental data along with the proposed model successfully accounts for the unique permeation characteristics of BCC metal membranes coated with catalytic layers and advances fundamental understanding of the permeation characteristics of the composite membrane, thereby accelerating the adoption and application of the composite membrane permeation model.

1. Introduction

Energy production from sustainable energy sources such as wind, solar and hydropower is highlighted as a promising method to reduce serious environmental problems [1]. The energy supply from these clean energy sources significantly varies due to seasonal and geographic reasons, and thus it is important to store surplus energy in a form that is easily transported by using chemical reagents, i.e., liquid organic hydrogen carrier (LOHC), ammonia, and formic acid [2–5]. An alternative energy carrier, namely hydrogen (H₂), is easily produced from the aforementioned chemical reagents by using a proper catalyst via a

dehydrogenation reaction and H₂ facilitates the production of electricity using H₂ fuel cells [6,7]. Generally, H₂-fuel cells need high-purity H₂ (>99.999%) with less than 1 ppm of CO. Therefore, it is important to develop efficient and continuous hydrogen separation and purification techniques for the upcoming 'hydrogen economy' [8]. A potential technique corresponds to membrane separation that does not require a regeneration procedure. This contrasts with conventional processes based on sorption, which requires regeneration routines for continuous hydrogen purification. Furthermore, a membrane reactor is also applied in various catalytic reaction processes to shift thermodynamic equilibrium via Le Chatelier's principle [9–11].

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