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A catalytic composite membrane reactor system for hydrogen production from ammonia using steam as a sweep gas

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ABSTRACT

For catalytic reactions involving H_2 extraction, the membrane reactor is an attractive option for enhancing the equilibrium and kinetics while eliminating excessive purification steps. In this study, a steam carrier adopted composite membrane reactor system is developed to produce pure H_2 (>99.99%) from ammonia with high H_2 productivity (>0.35 mol-H_2 g_{cat}^{-1} h^{-1}) and ammonia conversion (>99%) at a significantly reduced operating temperature (<723 K). Coupling of a custom developed palladium/tantalum composite metallic membrane and ruthenium on lanthanum-doped alumina catalysts allowed stable operation of the membrane system with significant mass transfer enhancement. Various reactor assemblies involving as-fabricated membranes and catalysts are experimentally compared to suggest the optimal configuration and operating conditions for future applications. Steam is adopted as a sweep gas, presenting efficient H_2 recovery (>91%) while replacing conventionally utilized noble carrier gases that require additional gas separation processes. The steam carrier presents similar membrane reactor performance to that of noble gases, and the water reservoir used for steam generation acts as an ammonia buffer via scrubbing effects. Finally, electricity generation is demonstrated using a commercial fuel cell along with process simulation, substantiating potential of the proposed membrane system in practical applications for H_2 production from ammonia and on-site power generation.

1. Introduction

The renewable energy share is growing as a mean to mitigate the global climate crisis and maintain sustainability. While renewable energy has great potential, its unpredictable nature presents challenges for direct integration into the grid. Additionally, either 1) it has a small energy production scale or 2) large production sites are too distant from energy utilization sites. Therefore, a large amount of renewable energy remains untapped, with limited logistics to store and transport the energy efficiently and economically. To resolve these temporal and spatial discrepancies between the supply and demand of renewable energy, energy carriers with high energy densities and economic feasibilities are being actively investigated.

Recently, ammonia has been identified as one of the energy carriers that allow efficient storage of renewable energy in a liquid form at a mild pressure under atmospheric conditions (0.8 MPa, 293 K [1]). When liquefied, it has a high energy density of 22.5 MJ kg^{-1} (higher heating value), with high gravimetric and volumetric H_2 contents of 17.6 wt% and 108 g L^{-1} at 293 K [2], respectively. Additionally, well-established storage and distribution infrastructure makes ammonia an economically feasible mass-scale energy carrier. While ammonia synthesis is energy-intensive, efforts have been directed toward making the process "greener" by adopting water electrolysis in conjunction with a conventional Haber Bosch process [3] or by electrochemical synthesis [4]. This

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