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Review Article

Perovskite-based proton conducting membranes for hydrogen separation: A review

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ABSTRACT

Hydrogen is considered a fuel of the future due to its diversified supply and zero greenhouse gas emission. The application of advanced membrane technology for hydrogen separation within the larger hydrogen production process context can substitute the use of more expensive and energy intensive cryogenic distillation and pressure swing adsorption technologies. This review overviews the basic aspects and progresses in perovskite-based proton conducting hydrogen separation membranes. Different configurations such as symmetric, asymmetric, hollow fiber, and surface modified perovskite membranes with various compositions are discussed and summarized. The challenges and future directions of such membranes are also elaborated.

Contents

Introduction 00
Perovskite structure 00
Transport mechanisms 00
    Hydration and proton concentration 00
    Intrinsic defects and doping 00
    Ambipolar transport and hydrogen flux 00
Current status 00
    Disk-shaped membranes 00

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Single-phase membranes . . . 00
Dual-phase membranes . . . 00
Asymmetric membranes . . . 00
Hollow fiber membranes . . . 00
Surface-modified membranes . . . 00
Challenges and outlook . . . 00
Limitations . . . 00
Future insights . . . 00
Conclusions . . . 00
Acknowledgment . . . 00
References . . . 00

Introduction

Over 60 million tons of hydrogen is produced per year for use in the chemical and refinery industries [1]. Hydrogen share in the energy market is increasing with the implementation of fuel cell systems and the growing demand for zero-emission fuels. Fuel cell electric vehicles (FCEVs) are powered by hydrogen and have the potential to revolutionize the transportation system. The northern German state of Schleswig-Holstein, for example, has declared plans to electrify its entire rail network by 2025, adopting fuel cell technology to power grids and also trains [2]. Plans are also in progress to realize hydrogen's potential in stationary applications. The United Kingdom City of Leeds has revealed its plan to become 'hydrogen city' in which the natural gas that runs through the city's pipes would be switched to hydrogen [3]. Japan has also announced plans to spend 40 billion yen to implement hydrogen technologies in the run up to the Tokyo 2020 Olympics [4]. As a part of the scheme, pipelines will be built that run through the athletes' village, delivering hydrogen to the industrial-scale fuel cells to power buildings [5].

Although hydrogen production technologies using renewable resources have been developed to achieve a sustainable energy cycle, hydrogen is still mainly produced from fossil sources due to technical and economical limitations of the existing technologies. In the near future, increased hydrogen production would likely be met *via* natural gas reforming [6]. Nevertheless, a large amount of carbon dioxide that is released during the hydrogen production from fossil fuels requires subsequent carbon dioxide capture and sequestration. Within this context, the development of hydrogen separation technologies remains one of the main spotlights that enables simultaneous high-purity hydrogen production and carbon dioxide capture. Pressure swing adsorption (PSA) and cryogenic distillation are currently being employed for hydrogen separation and recovery [7]. The PSA process relies on the adsorbents capacity to adsorb more impurities at high gas partial pressure than at low gas partial pressure. It offers the capability to produce very high purity hydrogen with up to 99.999% purity. Nevertheless, the PSA is only practical for medium and large industrial scale. The cryogenic distillation process is a sub-zero temperature separation process, which is based upon the difference in the boiling temperatures of the feed components. Due to its high capital cost, the cryogenic distillation is only suitable for continuous and large-scale operation. The purity levels of hydrogen are limited to 90–98% [8].

Advanced membrane technology, on the other hand, allows hydrogen recovery from low purity or low-pressure hydrogen containing stream that is uneconomical otherwise *via* PSA or cryogenic technologies. Its low capital cost, low energy consumption, and ease of operation make membrane technology attractive, even for low gas volume case. Polymeric membranes are usually made from glassy polymers (e.g., cellulose acetate and polysulfone), deriving their selectivity by discriminating between penetrants' components with subtle differences in size. They are used in the industry to recover hydrogen from hydrocarbon streams, such as refinery off-gases. Nevertheless, the polymeric membranes cannot withstand high temperatures, high pressures, and aggressive chemical environments. High pressures can cause compaction of the polymer that reduces permeability, or even the collapse of hollow fibers.

Inorganic membranes have in general higher flux performances than polymeric membranes that warrant their potential use for hydrogen separation at high temperatures, apart from their superior chemical and thermal stability. The inorganic membranes for hydrogen separation and purification can be classified into two main groups, i.e., (1) porous membranes such as silicas, zeolites, and carbon; and (2) dense membranes that comprise both dense and composite metal and ceramic membranes. Porous ceramic membranes possess high permeability, moderate to high selectivity, and are chemically and thermally stable. These qualities are attractive for hydrogen separation application. Porous membranes generally rely on molecular sieving effect of small pores (3–4 Å) to separate hydrogen from other gases (e.g., N_2, CO_2, etc.). Hydrogen fluxes through these membranes are promising when the operating temperatures are between 200 °C and 600 °C. Amorphous silica membranes have achieved reasonable combinations of hydrogen permeability and selectivity [9]. There are however many issues to be resolved before silica membranes can be fully applied such as the limited structure stability in atmospheres containing steam. Zeolite membranes have better thermal stability in the presence of steam than non-doped amorphous silica membranes due to their crystalline structure. Nevertheless, they have only moderate hydrogen selectivity given their large porous

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