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Hydrogen purification characteristics from ammonia decomposition gas with a plasma membrane reactor

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1. Introduction

Among the energy carriers, ammonia has attracted a lot of attention and has been actively studied and developed worldwide [(1)]. Ammonia has some advantages as an energy carrier. Therefore, the development of an on-site low-cost and high-efficiency hydrogen production system using ammonia is required for an energy system using ammonia as an energy carrier. Nowadays, thermal decomposition with a catalyst, such as nickel or ruthenium, is commonly used as dehydrogenation method for ammonia [(2)]. The dehydrogenation of ammonia with a catalyst only is not a suitable hydrogen production method for fuel cells because unreacted ammonia is mixed with the generated hydrogen. So, the hydrogen separation process is essential step to get pure hydrogen gas from various hydrogen careers. Therefore, a high-purity hydrogen production system comprising a catalytic decomposition reactor and a plasma membrane reactor (PMR) has been developed. In this system, ammonia is quickly decomposed into hydrogen and nitrogen in a catalytic reactor, and hydrogen is separated from ammonia decomposition gas by the PMR, high purity hydrogen (H_2 conc. = 99.99 %) is obtained continuously from ammonia. However, the hydrogen yield of this hydrogen production system was low (16 %). The cause of the low yield was the low hydrogen separation capacity of the PMR with ununiformed plasma firing.

The objective of this research is to develop PMRs packed with zeolite to improve the hydrogen production performance of the hydrogen production system. In this paper, the hydrogen purification properties of zeolite-loaded PMR, comparison of hydrogen purification properties according to zeolite species and feed gas composition, and hydrogen production properties of a device combining zeolite-loaded PMR and a catalytic reactor have been reported.

2. Experimental

Fig.1 shows H_2 purification experimental apparatus diagram. The experimental apparatus consists of a gas supply system, a high voltage pulse power supply, a plasma membrane reactor with zeolite (PMR), H_2 separation pump (N840.3FT.18ex, KNF). The PMR consists of a quartz tube (Outer diameter = 40 mm, thickness = 2 mm, length = 400 mm) and a H_2 separation membrane module, which are arranged coaxially. (Refer to the sectional view of Fig.1). The H_2 separation membrane module (manufactured by Nippon Seisen Co., Ltd.) is composed of three layers of a cylindrical H_2 separation membrane, a support and a cover (The support and the cover are made of SUS 316). The H_2 separation membrane is a Pd - 40% Cu alloy membrane (thickness = 20 μm). The H_2 separation membrane module doubles as an electrode on the high voltage side. The length of the gap between the quartz tube and the H_2 separation membrane module (called reaction gap) is 3 mm. A ground electrode (cupper mesh) is wrapped around the quartz tube. The length of the ground electrode is 300 mm, and the plasma is generated within the reaction gap inside the ground electrode. A columnar zeolite pellet (Tosoh Corporation) was filled in the gap inside the PMR. Atmospheric pressure plasma was generated in the reaction gap by dielectric barrier discharge (DBD) with a high voltage pulse power supply (manufactured by Sawafuji Electric Co., Ltd.). The flow rate of the test gas (H_2 100 % or H_2 75 %+N_2 25 %) was adjusted by a needle valve with a mass flow meter (MS-10SLPM, Alicat Scientific) and supplied to the PMR. H_2 in the test gas was separated and refined by the PMR.

3. Results and discussions

Fig. 2 shows the H_2 purification properties of PMR with zeolite. The results of PMR without zeolite are also shown as comparative data. The hydrogen purification performance was also improved by packing the zeolite in the PMR. The maximum H_2 purification flow rate obtained 205 L-H_2/h. The maximum H_2 purification rate of the PMR with zeolite improved from 16 % to 91 % compared to the PMR without zeolite. It has been reported that the field intensity of narrowest gap between dielectrics is greatly increased when plasma discharge is performed in a plasma reactor filled with a dielectric [3]. The high field intensity between dielectrics dissociates more hydrogen into protons in a short time, and proton permeation was promoted. In the PMR filled with zeolite, plasma discharge was also uniformly fired due to increase in field intensity.

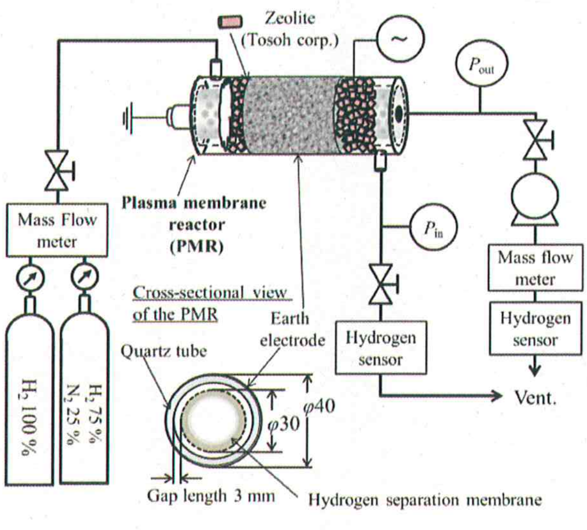

Fig. 1 Experimental setup of the hydrogen purification

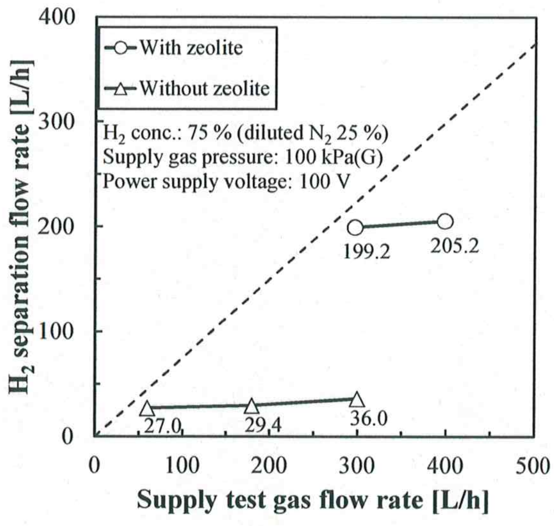

Fig. 2 The purification characteristics of the PMR with zeolite

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