NH₃ reforming by DBD using a H₂ permeable membrane

Yukio HAYAKAWA¹, Shinji KAMBARA¹, Tomonori MIURA²

 ¹ Gifu University, Environmental and Renewable Energy Systems Division, Graduate School of Engineering, 1-1 Yanagido, Gifu, 501-1193, Japan.
² Sawafuji Electric Co., LTD., 3 Nittahayakawacho, Ota, Gunma

 NH_3 is a H_2 storage material that may solve several problems related to H_2 transportation and storage in the H_2 society. Catalytic thermal decomposition is a promising technique for producing H_2 from NH_3 . This study investigated atmospheric plasma decomposition as a new H_2 production device. Therefore, it also observed that molecular NH_3 was rapidly decomposed by electron energy in the plasma and was converted into molecular hydrogen. The hydrogen production was increased by the NH_3 concentration, but H_2 conversion was dramatically decreased to 13.9 %, so unreacted NH_3 was existed. In order to improve these problems, we developed a new high voltage electrode which was equipped with a H_2 permeable membrane. At the result, this device could make high purity H_2 at room temperature and unreacted NH_3 was removed.

1. Introduction

Greenhouse effect gases are emitted from using various combustion resources like fossil fuel. As a result of present state, progress of the global warming is concerned about all over the world. In order to reduce greenhouse effect gas, the key is construction of the H₂ energy society, but H₂ has energy loss for its transportation and storages¹⁾. To resolve these problems, it is effective process that H₂ carrier accumulates and transports before it converts to H₂²⁾⁻³⁾. Particularly, NH₃ is expected as a H₂ energy carrier. NH₃ has a number of favorable characteristics, the primary one being its high capacity for H₂ storage, 17.6 wt%, based on its molecular structure. The secondary merit is that NH₃ itself is carbon–free at the end users, although CO₂ emission on production of NH₃ is dependent on the source of energy.

This research's objective is development of world first H_2 production devices integrating both of NH_3 and atmospheric plasma reactor to spread H_2 energy society.

In this paper, characteristics of NH_3 decomposition and H_2 production from NH_3 at the various conditions are reported.

2. Methodology

 NH_3 is decomposed H_2 and N_2 by electric discharge energy from atmospheric plasma (Eq. 1). But we expect that H_2 conversion is changed by various factors. This research evaluates possibility for practical use as new H_2 production devices.

 $NH_3 + e \rightarrow 0.5 N_2 + 1.5 H_2 + e$ (Eq. 1)

Fig. 1 shows experimental setup for H_2 production in a plasma reactor. First, NH_3 gas regulated mass flow controller is introduced into the plasma reactor, so NH_3 gas is decomposed in atmospheric plasma generated by dielectric barrier discharge. In these experiments, the gases decomposed and generated by atmospheric plasma are analyzed by gas chromatography (GC) and quadrupole mass spectrometer (Q–Mass), then H_2 conversion and H_2 production energy efficiency is calculated.

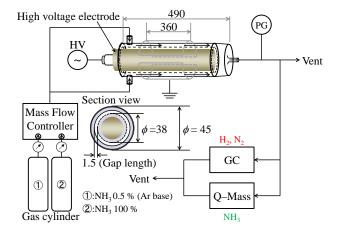


Fig. 1 Experimental setup for H₂ production.

3. Results

Fig. 2 shows effects of electric power and flow rates on H_2 conversion. In fig. 2, H_2 selectivity increased with the increasing of input power and decreased with the increasing of NH_3 gas flow rate. While H_2 selectivity was attained

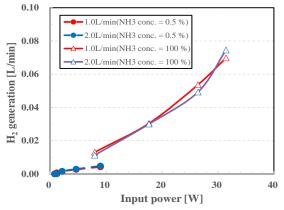


Fig.2 Comparison between low and high concentrations of NH₃ on H₂ generation.

100 % at low concentration NH_3 , H_2 selectivity was able to attain only 13.9 % at high concentration of NH_3 . So, it was clear that H_2 conversion was decreasing with an increase of NH_3 concentration.

Fig. 3 shows comparison between low concentration and high concentration on H_2 production. In fig. 3, H_2 production increased with an increase of NH_3 concentration. Based on fig. 2 and fig. 3, the higher NH_3 concentration had high performance of H_2 generation, but there was a problem of unreacted NH_3 because of low performance of H_2 conversion. While reforming the high NH_3 concentration gas, higher energy are added than the case of low concentration. Therefore, it thought the reverse reaction of generating NH_3 is in progress.

The inhibition of reverse reaction using the H_2 permeable membrane was tried in order to increase H_2 selectivity at high NH₃ concentration.

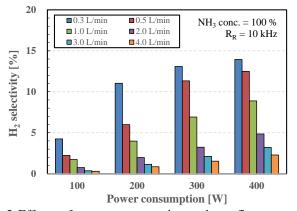


Fig.3 Effects of power consumption and gas flow rates on H_2 selectivity.

Fig.4 shows comparison experimental results between only plasma reforming and using permeable membrane at H_2 generation. In Fig. 4, it was found to be capable of continuous production of high purity H_2 at room temperature without a catalyst by mounting the permeable membrane to the plasma device. On the other hand, H_2 generation was improved by using a permeable membrane compared with results of NH₃ plasma reforming. Purity of generated H₂ was 100%.

Fig. 5 shows comparison energy efficiency between plasma reforming only and using permeable membrane. Energy efficiency is calculated for the input power. When permeable membrane is mounted, energy efficiency attained $6.5 \,$ %. Furthermore, the result of using permeable membrane is higher energy efficiency than the result of only plasma reforming. It was found that H₂ can obtain from NH₃ by introducing a permeable membrane to the plasma device.

4. Conclusion

At NH₃ 100 % gas, H₂ production is increased, but H₂ selectivity was reduced to 13.9% by reverse reaction of NH₃ generated occurs secondarily. To suppress reverse reaction, the hydrogen permeable membrane is mounted on the

plasma device. As the result, it is possible to suppress the progress of the reverse reaction as compared to before the permeable membrane mounted, H_2 generated flow rate is increased greatly. As a conclusion, it is considered to be capable of continuous production of high purity hydrogen at room temperature without a catalyst by using plasma device with H_2 permeable membrane.

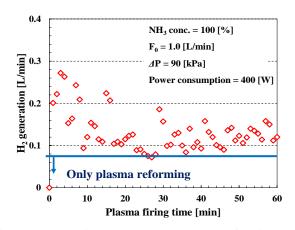


Fig. 4 Comparison between plasma reforming using permeable membrane and only plasma reforming on H_2 production.

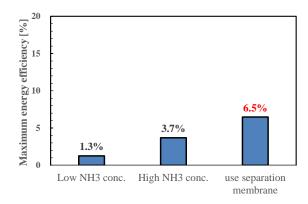


Fig. 5 Comparison between plasma reforming and using permeable membrane.

Referneces

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