Ammonia Decomposition Rate in Catalyst and Plasma

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Recently, in order to reduce emissions of CO_2, construction of hydrogen energy society is desired. However, the problem of hydrogen society is the major energy loss in the transportation and the storage of hydrogen. In order to solve this problem, it is effective to use ammonia which is a hydrogen-rich substance as a hydrogen carrier. This study aims to obtain high hydrogen generation rate from ammonia in catalytic thermal and plasma conditions. As results, the maximum hydrogen yield was achieved 98.4 % in catalytic thermal condition (NH_3 conc.= 30%, 850 ºC, 3600 /h), and 96.3 % in plasma condition (NH_3 conc.= 0.5 %, 0.2 L/min, 15 kV). And it was confirmed that H_2 production rate increased by using the H_2 separation membrane.

Keywords—ammonia decomposition, hydrogen production, catalyst, plasma

I. Introduction

In order to reduce emissions of CO_2, H_2 energy should be introduced to the transport sector and the consumer sector. The CO_2 emission of those sectors in Japan is about 46%. H_2 energy is very clean because H_2O is only generated when we consume H_2 energy, but H_2 has a large energy loss for its transport and storage[1)]. To resolve these problems, it is effective to transport and storage as H_2 carrier before it converts to H_2[2)]. NH_3 is especially expected as a H_2 energy carrier, because it has a number of favorable characteristics. The advantages of NH_3 as a H_2 energy carrier are that NH_3 has a high capacity for H_2 storage, 17.6 wt%, based on its molecular structure, and NH_3 itself is carbon-free at the end users.

We have developed a system for obtaining high-purity H_2 for fuel cells by atmospheric pressure plasma reactor with the high voltage electrode and the H_2 separation membrane after the thermal decomposition of NH_3 using catalyst. In this study, we have conducted experiments and simulations of NH_3 decomposition in the catalytic thermal and the plasma conditions.

II. Catalyst

(1) *Experimental setup and conditions*

Fig. 1 shows experimental setup for H_2 production in catalytic thermal condition. First, NH_3 gas was regulated by mass flow controller and introduced into the quartz tube (ϕ= 26 mm) in the gold furnace. So, NH_3 is decomposed and H_2 is generated by heat on the catalysts. In this experiment, we measured the N_2 and H_2 concentration of product gas by gas chromatography (GC), and the H_2 yield was calculated by Eq. 1.

$$H_2 \text{ yield } [\%] = \left\{ [H_{2,\,out}] \Big/ \frac{2 \times [NH_{3,in}] \times 0.75}{100 + [NH_{3,in}]} \right\} \quad \text{(Eq. 1)}$$

$[NH_{3,in}]$: inlet NH_3 conc.[%], $[H_{2,out}]$: outlet H_2 conc.[%]

In this experiment, we used Ni/Al_2O_3 catalysts (Ni=10 wt%) made in JGC Catalysts and Chemicals Ltd., and the shape of catalyst was ring. The inlet flow rate of NH_3 gas was regulated 1.0-3.0 L/min (space velocity; SV=3500-35000 /h) and reaction temperature was controlled 400-900 ºC. And the reaction mechanism was simulated by CHEMKIN-PRO.

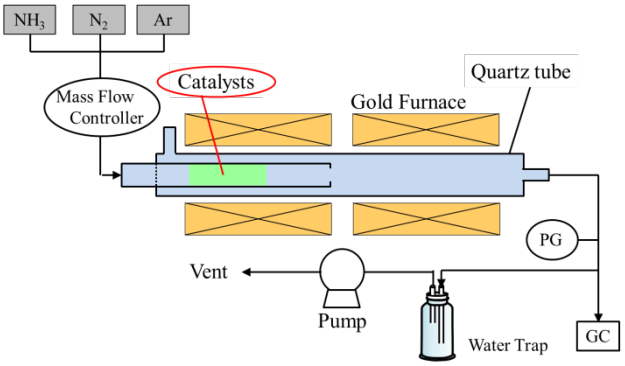

Fig. 1. Experimental setup for H_2 production in catalytic thermal condition

(2) *Result*

Fig. 2 shows the H_2 yield by using catalyst and blank for each reaction temperature and the simulation result when NH_3 conc.= 30%.

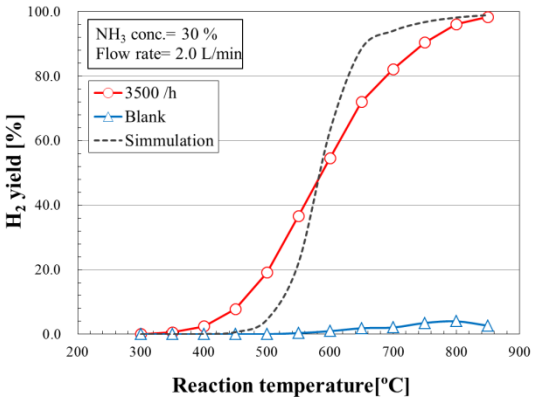

Fig. 2. H_2 yield for each reaction temperature in Blank test and SV=3500 /h and the simulation result when NH_3 conc. = 30%

In brank test, NH_3 was hardly decomposed, and H_2 was hardly generated. But, the H_2 yield increased by using Ni/Al_2O_3 catalyst, that is, we confirmed the active effect of Ni/Al_2O_3 catalyst. The H_2 yield increased as the reaction temperature increases. The maximum H_2 yield was 98.4 % (850 ºC). And simulation result almost matched the experimental result for SV= 3500 /h.

III. Plasma

(1) *Experimental setup and conditions*

Fig. 3 shows experimental setup for H_2 production in plasma condition. The NH_3 gas operation and measuring methods are almost same as catalytic thermal condition. In the plasma reactor, NH_3 is decomposed in atmospheric plasma generated by dielectric barrier discharge.

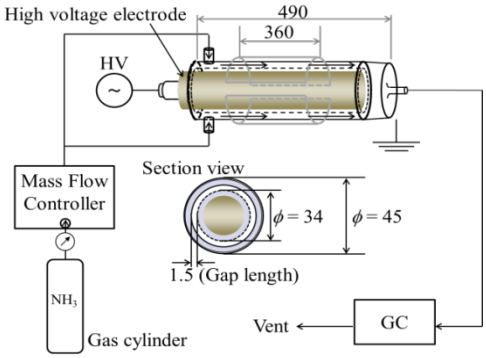

Fig. 3. Experimental setup for H_2 production in plasma condition

In this experiment, we used the low concentration NH_3 gas(= 0.5 %) because it was assumed that the NH_3 gas was decomposed by using catalyst. The inlet flow rate of NH_3 gas was regulated 0.2-2.0 L/min and the applied voltage was controlled 3.5-15.0 kV.

(2) *Result*

Fig. 4 shows the H_2 yield for applied voltage in various flow rate when NH_3 conc.= 0.5 % and simulation results.

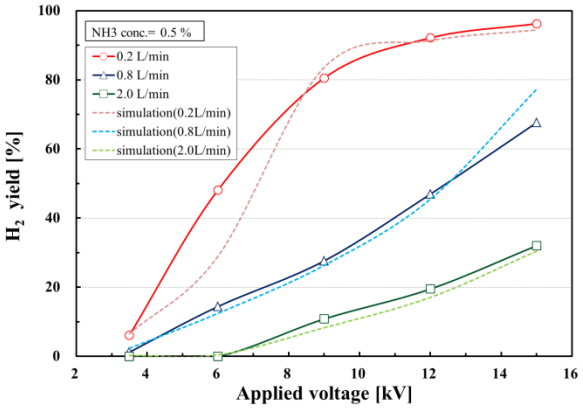

Fig. 4. H_2 yield for applied voltage in various inlet gas flow rate when NH_3 conc. = 0.5 %

It was found that H_2 yield increased as the flow rate decreases or the applied voltage increases. The maximum H_2 yield was 96.3 % when the flow rate is 0.2 L/min and the applied voltage is 15.0 kV. And simulation results almost matched the experimental results.

The results as experiments, it was found that almost NH_3 was decomposed by atmospheric pressure plasma reactor with the high voltage electrode and the H_2 separation membrane after the thermal decomposition of NH_3 using catalyst. But high-purity H_2 for fuel cells is needed more low concentration of remaining NH_3. So, we introduced the H_2 separation membrane to obtain high-purity H_2 for fuel cells. Fig. 5 shows H_2 production rate by the H_2 separation membrane for each applied voltage when NH_3 conc.= 100 %.

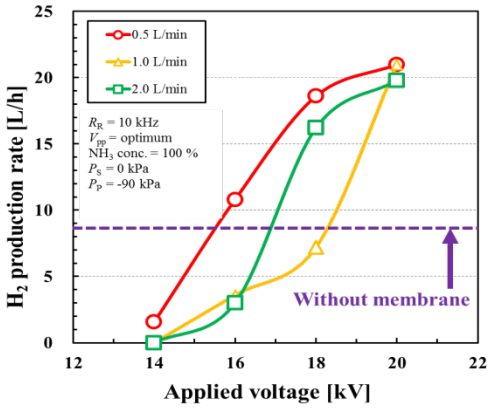

Fig. 5. H_2 production rate by the H2 separation membrane for each applied voltage when NH3 conc.= 100 %.

By using the H_2 separation membrane, we were able to obtain a lot of the high-purity H_2 more than the result without membrane.

V. Conclusion

H_2 energy is very clean, but H_2 has a large energy loss for its transport and storage. So, it is effective to transport and storage as H_2 carrier. NH_3 is especially expected as a H_2 carrier. We have developed a system for obtaining high-purity H_2 for fuel cells by atmospheric pressure plasma reactor with the high voltage electrode after the thermal decomposition of NH_3 using catalyst.

In catalytic thermal condition, the maximum H_2 yield was 98.4 % (NH_3 conc.= 30 %,850 ºC). In plasma condition, the maximum H_2 yield was 96.3 % (NH_3 conc.= 0.5 %, 0.2 L/min, 15.0 kV). And it was confirmed that a lot of the high-purity H_2 was obtained by using the H_2 separation membrane more than the result without membrane.

References

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