Effect of pressure range on ammonia synthesis using pressure swing in N_2-H_2 non-thermal plasma

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Abstract: In this contribution, we report a novel ammonia synthesis process in which the pressure of N_2-H_2 non-thermal plasma system swings to improve ammonia synthesis efficiency. We succeeded in fabricating a plasma reactor with a compression ratio of 10 and were able to confirm the improved synthesis efficiency under the pressure swing condition.

1. Introduction

Ammonia is an extremely important compound industrially, and its importance has increased further in recent years as it has been considered for use as a hydrogen carrier and alternative fuel to fossil fuels. In our laboratory, we are developing a novel process for synthesizing ammonia by generating plasma through pressure swings in a reactor[1]. In a previous study, it was confirmed that the synthesis efficiency can be improved by generating plasma while swinging the pressure. However, the compression ratio of the reactor was as low as 3, and increasing the compression ratio was an issue for improvement. In this study, a reactor with a compression ratio of about 10 was fabricated and we will conduct experiments over a range of pressures in order to find the optimal pressure swing conditions and explore the reaction mechanism of ammonia synthesis using plasma in order to improve the synthesis efficiency.

2. Methods

A schematic diagram of the experimental apparatus is shown in Figure 1. The reactor consists of a quartz glass cylinder with an inner diameter of 26 mm, a 2 mm thick TEMPAX glass as a cylinder head, an aluminum piston, and inlet/outlet solenoid valves. Pressure swing was realized and controlled by synchronizing the motion of piston and the opening/closing timing of the solenoid valves. The surface discharge and/or space discharge were created on the surface of cylinder head glass and/or in the space between the cylinder head and the piston crown, respectively. The backside of the cylinder head glass was covered by high-voltage copper electrode which was connected to a high-voltage transformer. Ammonia synthesis was performed using 10 sccm of H_2 and 10 sccm of N_2 as feedstock, and the product gas was trapped by bubbling in a 4 mmol/L methanesulfonic acid solution, and the amount produced was measured using HPLC.

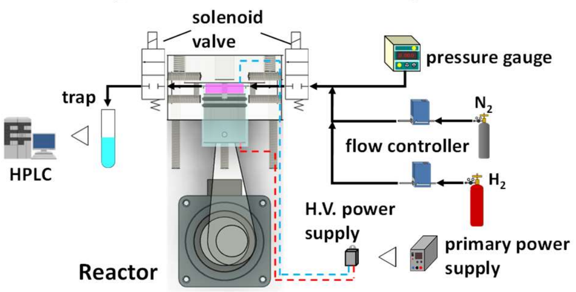

Fig. 1. experimental apparatus

3. Results and Discussion

Ammonia synthesis was performed under constant pressure and pressure swing conditions, and results were compared in terms of energy efficiency. The results are shown in Figure2. Under the constant pressure condition, the energy efficiency was significantly higher on the low pressure side, which is thermodynamically unfavourable for ammonia synthesis but kinetically preferable for the dissociation of nitrogen molecules by electron impact reactions. On the other hand, under the pressure swing condition, the energy efficiency was higher than that under the constant pressure condition in all pressure swing ranges although no significant differences in the results were observed between the pressure swing ranges. This is considered to be due to the balance between the kinetic positive effect at low pressure and the thermodynamic positive effect at high pressure. Therefore, it is considered important to synchronize the discharge and pressure swing and optimize the discharge timing.

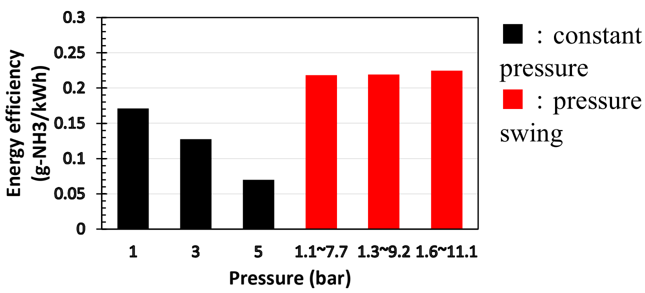

Fig. 2. Synthesis efficiency of ammonia in different pressure ranges

4. Conclusion

A reactor with a compression ratio of 10 was successfully fabricated. By comparing the constant pressure condition and the pressure swing condition, we confirmed the improved synthesis efficiency under the pressure swing condition. It is thought that synchronization of the discharge and pressure swing conditions will make the effect of the pressure swing at high compression ratios more effective.

References

[1] Mori, S. *et al.*, J. Chem. Eng. Japan, **53**, 498 (2020)