Modeling of CO_2 conversion in non-thermal plasma for high conversion and energy efficiency

Shinsuke Mori, Kumpyo Kwak, Kai Fukunaga, Yuki Yokoyama
Department of Chemical Science and Engineering, Institute of Science Tokyo, Tokyo, Japan

Abstract: In this contribution, we develop a simple calculation model to estimate carbon dioxide conversion in various carbon dioxide discharge systems by combining partly analytical solutions and numerical calculations and verify the applicability and demonstrate the effectiveness of the created model by comparing it with experimental data on carbon dioxide decomposition in various discharge systems.

1. Introduction

The importance and attention given to plasma-based CO_2 decomposition processes has increased dramatically due to the growing focus on the recycling of carbon dioxide using renewable electricity. Although a great number of experimental and computational results have been reported, neither experimental nor computational reproduction of the extremely high energy efficiencies of over 80% reported in the 1980s has yet to be achieved. One of the reasons for these discrepancies in experimental and computational results can be attributed to incorrect estimates of dissociation reactions via vibrationally and electronically excited states, but the details have not yet been fully elucidated. Therefore, this study aimed to develop a model that can reproduce the results of carbon dioxide decomposition by various plasma systems in a unified and simple manner.

2. Methods

We attempted to develop a simple calculation model to estimate carbon dioxide conversion in various carbon dioxide discharge systems. For that purpose, a calculation model combining partly analytical solutions and numerical calculations was developed and compared with not only the newly acquired experimental data but also our previous experimental data [1,2].

3. Results and Discussion

Figure 1 compares experimental results and modelling results. The experimental data were obtained from a carbon dioxide dissociation experiment using a DC glow discharge in a discharge tube with an inner diameter of 3 mm. This experiment is characterized by the fact that carbon dioxide is decomposed using only the part of the positive column along the discharge tube where the electric field intensity and electron density are considered to be constant, which is useful for comparison with calculations. In the calculations here, the rate of conversion of carbon dioxide is obtained by assuming plug flow, using the analytical solution of a first-order reversible reaction with the electron impact dissociation reaction of carbon dioxide as the rate-limiting step, and numerically analyzing only the rate constant and electron density used in the analytical solution. Since the reduced electric field is known to be 22 Td from the experimental data, the rate constant for electron impact dissociation was calculated with Bolsig+ [3] using experimentally derived reduced electric field [1].

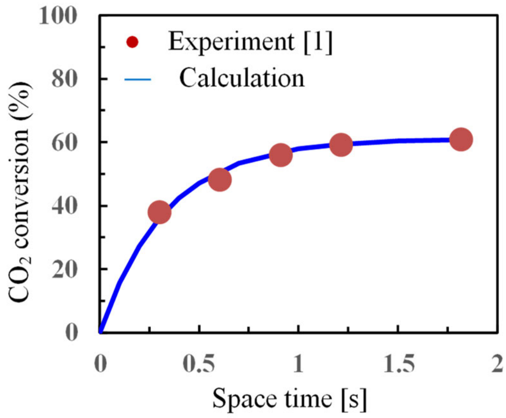

Fig. 1. Comparison between experimental results and calculation results for CO_2 conversion. (I = 4.0mA, E/N = 22Td)

The cross-section data for carbon dioxide dissociation recommended in [4] was used in this calculation. The electron density was also calculated using the diameter of the discharge tube, the current value, and the mobility calculated with Bolsig+. As a result, the calculated results reproduce the experimental results with very good accuracy, even though the rate constant for the reverse reaction, 1.1 (1/s), is the only parameter used in the fitting. The applicability of the model was verified using the same approach by using experimental data in various discharge systems.

4. Conclusion

A simple computational model combining analytical solutions and numerical calculations was developed and used to verify the applicability and demonstrate the effectiveness of the created model by comparing it with experimental data on carbon dioxide decomposition in various discharge systems.

References

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