ロングDCアークを用いたCF4分解による温室効果の削減 Reduction of greenhouse effect by CF4 decomposition using long DC arc

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1. Introduction

Carbon tetrafluoride, CF₄, is mainly used as the dry etching gas in semiconductor industry. However, it has a high global warming potential about 6,500, and is difficult to be decomposed. Abatement processing with thermal plasmas has been implemented in industrial fields due to their advantages of high temperature and high chemical activity. Long DC arc has a long electrode gap distance of 300 mm, resulting in sufficiently long residence time for decomposition of harmful target [1]. The purpose of this study is to decompose CF_4 by long DC arc and to investigate decomposition mechanism.

2. Experiment

The setup consists of a power supply, a plasma torch, and a scrubber shown in **Fig. 1**. The arc current was 10 A. Nitrogen at 25 L/min was used as the plasma gas, while CF₄ was injected at 0.5 L/min. A decomposition experiment was conducted under two conditions; i) hydrogen addition up to 2.0 L/min and ii) steam addition up to 2.0 L/min. Generation of H, O and OH radical by the additional gas inhibit recombination of CF₄ outside the discharge region.

The produced gases were analyzed by a gas chromatograph (GC) and a quadrupole mass spectrometer (QMS) to investigate the destruction and removal efficiency (DRE) and the composition of the produced gases.

3. Results and Discussion

Figure 2 shows the relationship between the H/F molar ratio and DRE. The DRE was 0% at H/F=0. This is because C and F radicals are easily recombined to CF_4 after thermal plasma decomposition. The DRE value increased with an increase of H/F ratio. The maximum DRE value was 95% at H/F=2.0. High flow rate of steam enhances oxidation atmosphere, therefore C and O recombine to form stable CO and CO₂. In addition, highly reactive F radicals are recovered by forming HF. This leads to suppression of CF₄ recombination.

The recombination mechanism after plasma decomposition is discussed here on the basis of the temperature dependence of Gibbs free energy change. Recombination temperature of HF is 4,740 K, while

that of CF is 4,670 K. This small difference of recombination temperature suggests that the recovery of F radicals by H is difficult due to the competitive reaction of CF recombination. In contrast, the recombination temperature of CO is about 7,600 K. This strongly suggests that the oxidation of C radicals is a key reaction to suppress CF_4 recombination.

4. Conclusion

Carbon tetrafluoride was successfully decomposed by long DC arc with steam addition. Decomposition and recombination mechanism of CF_4 in long DC arc was clarified. Long DC arc system is expected to play an important role in the semiconductor industry to reduce greenhouse gas emissions.

References

[1] S. Choi, et al., J. Chem. Eng. Jpn, 46, 201-208 (2013).



Fig. 1 Schematic diagram of long DC arc system.



Fig. 2 Effect of H/F ratio on the DRE, additional hydrogen and steam.