Formation of stable species in a coaxial DBD with short residence times in argon-tetramethylsilane: comparing experiment and 1d-t fluid modeling

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Abstract: Infrared-spectrometric density measurements of stable species generated in a coaxial dielectric barrier discharge operated in ambient-pressure argon with small admixtures of tetramethylsilane are reported and compared with results of time-dependent, spatially one-dimensional fluid modeling. For the considered residence times up to 8 ms, the agreement is significantly better than an order of magnitude for five out of seven species studied.

1. Introduction

The investigation of plasma-chemical and -physical processes in dielectric barrier discharges (DBDs) at atmospheric pressure fed with small amounts of organosilicons in rare gases has gained increasing interest for plasma-enhanced chemical vapor deposition during the last decades. To get a deeper understanding of the plasma chemistry in Ar discharges with small fractions x_M of tetramethylsilane (TMS), Fourier-transform infrared (FTIR) spectroscopy has been applied to analyze the formation of stable molecules at the outlet of a coaxial DBD for different residence times. The measurements have been accompanied by numerical modeling employing a time-dependent, spatially one-dimensional (1d-t) fluid-Poisson model. First results of the comparative investigations are depicted and discussed.

2. Methods

The symmetric, coaxial DBD reactor consists of two borosilicate glass tubes with 2-mm wall thickness and a gap of 1 mm. On the outside, a 2 mm wide aluminum adhesive tape serves as grounded electrode. The high-voltage electrode in the inner tube consists of copper powder and is also 2 mm wide. The discharge is powered by a sinusoidal voltage (4 kV amplitude, 86.2 kHz). Gas residence times t_r in the discharge between 1 and 8 ms were obtained by varying the average gas flow velocity v_{av}. Effluent analysis was performed using an FTIR spectrometer equipped with a long-path (16 m) gas cell.

The modeling studies were done using a 1d-t approach similar to that presented in [1]. It includes an extensive plasma-chemical model for Ar-TMS mixtures taking about 90 species and 700 reactions into account.

3. Results and discussion

Figure 1 compares measured number densities of stable molecules with modeling results for an Ar plasma with an initial TMS admixture of 100 ppm. The depletion of TMS during its residence time in the plasma zone leads to the formation of various silicon-containing species as well as hydrocarbons. Trimethylsilane [$(CH_3)_3SiH$] is the predominant silicon-containing molecule generated in the discharge. In addition, large amounts of methane (CH_4), ethane (C_2H_6), ethylene (C_2H_4) and acetylene (C_2H_2) are found. The experimental and modeling results generally show similar trends and, partly, agree well. Notice that the number density of hexamethyldisilane [$(CH_3)_3SiSi(CH_3)_3$] was not accessible experimentally.

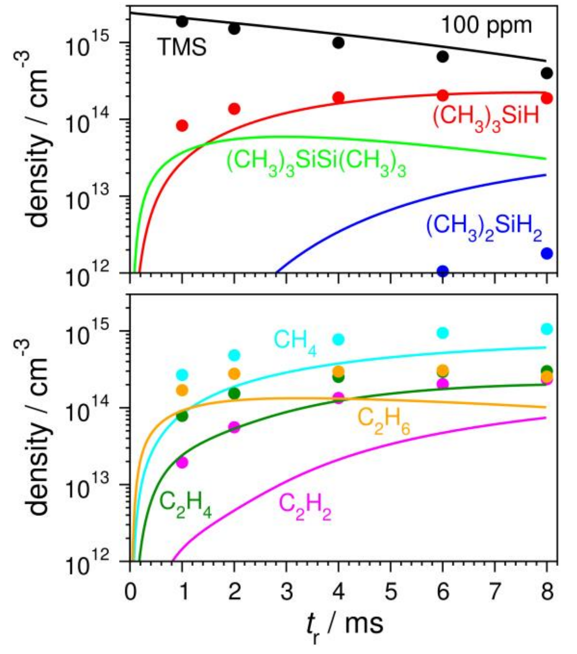

Fig. 1. Number densities of stable molecules as function of the residence time t_r from FTIR measurements (symbols) and modeling (lines) for 100 ppm TMS

4. Conclusion

Number densities of stable molecules formed in ambient-pressure coaxial DBDs in Ar-TMS are quantified by FTIR spectroscopy. The general agreement between experimental and modeling results confirms the performance of the model. Discrepancies for individual species help to improve the kinetic parameters applied.

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References

[1] D. Loffhagen et al., Plasma Chem. Plasma Process., **41**, 289-334 (2021).