FORMATION OF POLYMER PARTICLES AND GASEOUS PRODUCTS FROM CS2 IN A NON-THERMAL PLASMA IN AIR

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Abstract

Experiments and numerical calculations have been performed to investigate kinetic mechanisms of CS2 oxidation and formation of polymer particles in a non-self-sustained glow discharge in air. Both mechanisms of CS2 conversion were realized in two different experimental setups. In the first case conversion occurred with formation of carbon and sulfur oxides, and in the second case solid polymer particles arose.

1. Introduction

Carbon disulfide impurities can be found in toxic wastes of some chemical companies. To optimize CS2 decomposition in air, it is necessary to study the most efficient mechanisms of plasma chemical conversion of CS2 vapors. It is known that the CS2 conversion in the presence of oxygen may occur following different ways. If the e-beam produces mainly free radicals, such as O or O1, then a chain oxidation of CS2 is realized. The main products of this process are sulfur powder, carbon and sulfur oxides. With more intensive creation of positive and negative ions the CS2 molecules are condensed in the form of polymer particles [1].

The objective of the present work is to study the processes of the CS2 conversion in N2/O2/CS2 mixtures irradiated by electron beams.

2. Experiment and modelling

In the experiment the both mechanisms of carbon disulfide conversion were realized using e-beam accelerators of two different types: with an e-beam of current density 800 A/cm2 and duration of 3 ns; with an e-beam of current density 10^{-3} A/cm2 and duration of 48 μs. In both systems dc voltage was applied to the discharge gap to investigate the influence of the electric field on the energy consumptions for CS2 conversion. To detect gaseous products of the CS2 conversion gas chromatography methods were used. The experimental setup is described elsewhere [2]. To supplement the experiment, numerical simulation of the kinetics processes occurring in the gas discharge plasma was performed. The model of a non-self-sustained discharge included simulations of an electron beam with allowance made for the secondary electron contribution to the excited states of molecules, circuit for powering the discharge gap, plasma parameters (conductance, electron temperature), rate constants for electron-collision elementary processes, and the kinetics for free radicals. The spatial distribution of all particles in the plasma was considered uniform. The set of reactions comprised the processes where electrons, neutral particles, positive and negative ions participated and took into account of 300 reactions.
3. Results

With the first setup (high current e-beam), the specific energy consumptions for \( \text{CS}_2 \) removal from air were of 0.6 eV per molecule and in second setup - 0.3 eV. The numerical simulation has shown, that the low energy consumptions for high current e-beam are due to the chain mechanism initiated by O radicals:

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\begin{align*}
\text{CS}_2 + O & \rightarrow \text{CS} + \text{SO} \\
\text{CS} + O & \rightarrow \text{CO} + \text{S} \\
S + O_2 & \rightarrow \text{SO} + O \\
O + \text{CS}_2 & \rightarrow \text{COS} + S \\
O_2 + \text{CS} & \rightarrow \text{CO} + \text{SO} \\
O + \text{NO}_2 & \rightarrow \text{NO} + \text{SO} \\
\text{SO} + O_2 & \rightarrow \text{SO}_2 + O \\
2\text{SO} & \rightarrow \text{SO}_2 + S \\
S + \text{CS}_2 & \rightarrow \text{CS} + \text{S}_2 \\
\text{SO} + \text{NO}_2 & \rightarrow \text{NO} + \text{SO}_2 \\
O + \text{S}_2 & \rightarrow \text{SO} + S \\
\text{SO} + \text{CS} & \rightarrow \text{CO} + \text{S}_2 \\
\text{COS} + O & \rightarrow \text{CO} + \text{SO} \\
O + \text{CS}_2 & \rightarrow \text{CO} + \text{S}_2
\end{align*}
\]

The above reaction set shows that main products of \( \text{CS}_2 \) decomposition are carbon and sulfur oxides. Numerical calculation has shown that concentration of charged particles \( n_0 \), in this case is two orders lower than concentration the free radicals. The efficiency of the oxidative conversion of \( \text{CS}_2 \) is influenced by vibrationally excited states of \( \text{N}_2^* \) due to the reaction \( O + \text{N}_2^* \rightarrow \text{NO} + \text{N} \), which eliminates concentration of O radical and results in increasing NO.

The ion density in the plasma produced by the microsecond e-beam of current density \( 10^4 \text{ \text{A/\text{m}^2}} \) is comparable to that of free radical. Because the nucleation processes are more intensive for ions than those of oxidation with participating of free radicals the polymerization processes mainly define the decrease in the carbon disulfide concentration. At the same time, the fraction of ions lost due to electron-ion recombination decreases steeply since the electron density in the plasma is two orders of magnitude lower. Favorable conditions for the formation of condensation centers are thereby established in the plasma. The ion-molecular processes, which bring about the formation of active centers, develop rather intensively [3] and hence the rate of the \( \text{CS}_2 \) removal from air by the microsecond low-current beam substantially increases (fig. 1).

4. Conclusions

The main channels for the conversion are the carbon disulfide oxidation following the chain mechanism with participation of free radicals and formation of solid particles. Separate experiments have been performed for each of these mechanisms. The low energy consumption for removing one \( \text{CS}_2 \) molecule, which has been obtained in the experiments, are explained by the chain character of the processes occurring in the plasma. The results obtained are of interest in the technologies of air purification from toxic impurities.
Fig. 2. Time dependencies of the concentrations of the charged particles produced by a nanosecond (solid curves) and a microsecond (dash curves) e-beams.

References
