

Processes of carbon disulphide conversion in pulsed corona discharge plasma

D.L. Kuznetsov, I.E. Filatov, V.V. Uvarin, Yu.S. Surkov, S.A. Nikiforov, G.G. Ugodnikov

*Institute of Electrophysics of the Ural Branch of the Russian Academy of Sciences,
Ekaterinburg, Russian Federation*

Carbon disulphide CS_2 is a toxic component of exhaust gases from cellophane and viscose production. In the mid-90s research of CS_2 conversion in plasma formed by pulsed electron beams and non-self-sustained discharges initiated by electron beams was started [1]. It was found that the conversion mechanism and its products are dependent on the parameters of the beams and discharges. The conversion is carried out with a low specific energy expenditures. However, the widespread use of electron beams in actual air treatment systems is constrained by three facts: the complexity and cost of the electron-beam systems, lack of reliability, as well as the need to create a biological protection against ionizing radiation. The more prevalent methods are frequently used, such as streamer corona, because such installations are relatively cheap, reliable, easy to operate and do not require the creation of biological protection against ionizing radiation. There are reports on the use of the corona discharge at a constant voltage for the conversion of carbon disulphide [2].

We propose to use a rep-rated streamer corona with nanosecond pulse duration to study conversion of carbon disulphide in air.

For experiments, the installation has been created with parameters presented in [3]. A cylindrical reaction chamber with a 0.24 mm diameter nickel wire on its axis serves for gas mixture processing. The nickel wire is a cathode of streamer corona discharge. A hollow stainless steel 88 mm diameter cylinder arranged inside the chamber is an anode of the discharge. High voltage pulses are applied to the cathode from the output of generator SM-4N. Pulses of voltage and current are controlled via four-channel digital storage oscilloscope Tektronix TDS5054 with a 500 MHz bandwidth. Absolute CS_2 and CO_2 concentrations and relative sulphur dioxide SO_2 and carbonyl sulphide COS concentrations were controlled with the use of the gas chromatograph “Tsvet-500M”. Streamer corona parameters were as follows: voltage pulse 150–170 kV, discharge current pulse 100–300 A, pulse duration 15–30 ns, pulse repetition rate 1–10 Hz. A mixture of 50–10000 ppm of CS_2 and of atmospheric pressure air was investigated. View of the installation is shown in Fig. 1.

It turned out that the main products of the conversion were SO_2 , COS and CO_2 .



Fig. 1. View of the installation

Fig. 2a shows dependencies of the decrease in CS_2 and the increase in SO_2 and COS concentrations on the number of discharge shots for initial CS_2 concentration of 10000 ppm.

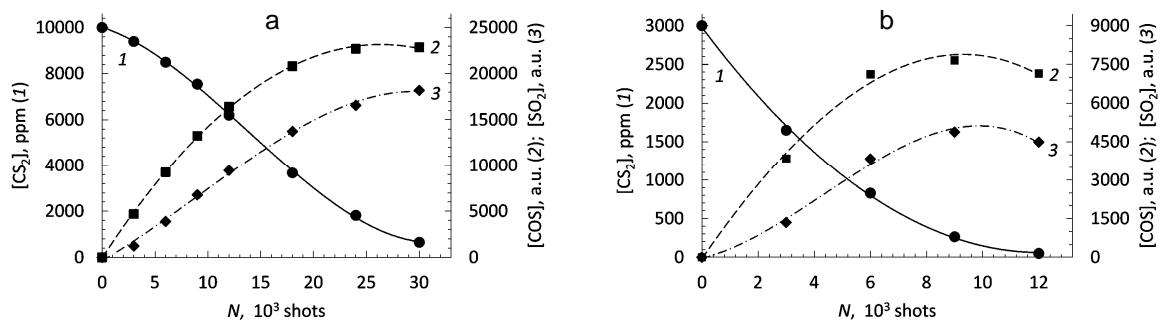


Fig. 2. Dependencies of CS_2 (1), COS (2) and SO_2 (3) concentrations on the number of streamer corona shots N for initial mixture composition 10000 ppm CS_2 (a) and 3000 ppm CS_2 (b) in atmospheric pressure air.

Pulse repetition rate $F = 5\text{Hz}$

In the range of CS_2 concentration 10000–4000 ppm this concentration decreases almost linearly through N . It is the case of zero order of reaction by concentration, when there is excess of the main reagent. Further, a curve slope reduction is observed in the range of CS_2 concentration less than 4000 ppm. It is the case of first order of reaction by concentration, when there is low concentration of the main reagent. Almost a linear increase in COS and SO_2 concentrations is observed during first 12000 streamer corona shots. Further, slowdown of COS and SO_2 concentrations occurs, and a saturation is observed after 25000 shots. Since SO_2 and COS are also destroyed under effect of streamer corona, the saturation indicates that the rates of formation and destruction of these products compared. It was determined that after 30000 shots concentration of CS_2 was reduced to 650 ppm with cleaning degree 93.5% when initial CS_2 concentration was 10000 ppm. Energy efficiency of CS_2 removal was 360 $\text{g} \cdot (\text{kW} \cdot \text{h})^{-1}$, and specific energy expenditures for the conversion were $\sim 8 \text{ eV/molecule}$.

We can also estimate upper limit of SO_2 and COS concentrations. Since one CS_2 molecule may produce two SO_2 molecules, or two COS molecules, or one SO_2 and one COS molecule,

the maximum SO_2 and COS concentration in Fig. 2a cannot exceed in total of 20000 ppm (2%). As the figure shows that the concentration of these products are comparable, it is likely that the concentration of each of the products does not exceed 10000 ppm (1%).

Fig. 2b shows dependencies of the decrease in CS_2 and the increase in SO_2 and COS concentrations on the number of discharge shots for initial CS_2 concentration of 3000 ppm. In all range of CS_2 concentration 3000 ppm – 50 ppm $[\text{CS}_2]$ decreases exponentially through N . It corresponds to first order of reaction by concentration. A monotonous increase in $[\text{COS}]$ and $[\text{SO}_2]$ occurs during first 6000 shots. Further, slowdown of $[\text{COS}]$ and $[\text{SO}_2]$ takes place, and a decrease in their concentrations starts after 9000 shots. Since $[\text{CS}_2]$ becomes close to zero after 10000 shots, the formation of COS and SO_2 stops, and their decomposition by streamer corona continues. That is why the products concentrations are reduced after 10000 shots. In this experiment concentration of carbon disulfide was reduced to 50 ppm, i.e. 98%, after 12000 shots. Energy efficiency of CS_2 removal was $260 \text{ g} \cdot (\text{kW} \cdot \text{h})^{-1}$, and specific energy expenditures for the conversion were $\sim 11 \text{ eV/molecule}$. After spending arguments similar to those in the discussion of Fig. 2a, it can be assumed that the maximum concentrations of SO_2 and COS in Fig. 2b will likely not exceed 3000 ppm each.

The obtained values of specific energy expenditures for CS_2 conversion were higher than in the case of the use of pulsed electron beams [1], but less than the typical values of specific energy expenditures for CS_2 conversion using electric discharge methods [2].

It was determined that when the mixture treatment was continued after decomposition of the bulk of CS_2 , a destruction of SO_2 and COS occurred. In this case, the fixed final products of CS_2 conversion were sulphuric acid H_2SO_4 and carbon dioxide CO_2 . Sulphuric acid is formed by reacting sulphur oxide SO_3 with water, because water vapor is always present in the air. It was reported that during CS_2 conversion under the influence of pulsed electron beams and non-self-sustained discharges, initiated by electron beams, a conversion mechanism similar to chain one of CS_2 combustion in oxygen [1]. Main reactions of the mechanism are





Initiation of the process of CS_2 oxidation occurs in reactions of dissociative electron attachment to O_2 (1) and CS_2 (2). With sufficient amount of O_2 full CS_2 oxidation is described by reaction



According to the presented model main products of CS_2 conversion are SO_2 , COS and CO_2 . Depending on concentration of active particles in plasma this mechanism may be realized as chain (with repeated participation of formed particles in reactions of the mechanism) or non-chain. A difference should mainly be not in reaction products set but in energy efficiency of conversion process.

According to the results of our experiments and evaluations it was revealed that the mechanism of plasma-chemical and chemical reactions of CS_2 conversion as a whole is the same for the plasma produced by an electron beam, non-self-sustained discharge and streamer corona. The main difference is that in the plasma produced by a high-current nanosecond electron beam higher concentrations of charged, excited particles and radicals are achieved than in the plasma produced by a nanosecond streamer corona. As a result, in the beam plasma it is possible to implement the conversion of CS_2 by a chain mechanism with specific energy expenditures of less than 1 eV/molecule, and in a streamer corona plasma, under the same reactions and products, the chain mechanism is not performed, which leads to an increase in energy expenditures for the conversion. However, the main products, with a few exceptions, are the same.

Thus, these experiments allow us to consider the pulsed corona discharge as a new effective method for the conversion of carbon disulphide in the air. The efficiency of this method is slightly lower than when using the pulsed electron beams, and non-self-sustained discharges initiated by the electron beams [1], but significantly higher than when using a DC corona discharge [2]. In addition, the advantages of this method compared with the electron-beam one are simplicity of construction, low cost, high reliability and security.

The work was supported by Russian Foundation for Basic Research (project No 12-08-00130-a).

References

- [1] Y.N. Novoselov, A.I. Suslov, D.L. Kuznetsov, Rus. Technical Physics 48(6) (2003) 780-786.
- [2] X. Yan, Y. Sun, T. Zhu, X. Fan, Journal of Hazardous Materials 261 (2013) 669-674.
- [3] Kuznetsov D.L., Filatov I.E., Surkov Yu.S., Izvestiya Vuzov. Fizika 55(10/3) (2012) 236-240.