Oxidation of an acetaldehyde/acetylene equimolar mixture by an atmospheric non-thermal plasma discharge

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Abstract: The present work has highlighted the good efficiency of an atmospheric plasma corona discharge working in a pulsed regime to remove 500ppmC of acetylene and 500 ppmC of acetaldehyde, two model molecules of engines exhausts, both diluted in a N_2 gas flow containing 5% of O_2. This efficiency was characterized by a removal of 99% of the initial 500 ppmC of acetaldehyde and of 66% of the initial 500 ppmC of acetylene for a specific input energy inferior to 200 J/L. However the global selectivity in CO and CO_2 is quite low, not raising over 30% of the carbon converted. Simulations performed by using a quasi-homogenous model, have showed that acetaldehyde is mainly destroyed during the pulse time while acetylene is mainly destroyed by oxygenated species (O, OH) during the post-discharge time.

Motivation

Among the air and industrial exhaust gas possible treatments, atmospheric plasma processes have been the subject of many studies for the last decade and have demonstrated a quite good efficiency for the removal of an unique pollutant molecule [1]. Nevertheless studies concerning the common removal of several pollutants by those atmospheric discharges are still scarce. This work propose to focus on the availability of an atmospheric pulsed discharge to remove 1000 ppmC of acetaldehyde and acetylene, both well known representative molecules of volatile organic compounds emitted in the exhaust gas.[2].

Experimental part

The pulsed atmospheric plasma discharge is generated by a Marx generator allowing to reach a 20-30kV voltage on the cathode. The gas inlet is mainly composed by a N_2/O_2 mixture containing 5% of oxygen added with 1000 ppmC of pollutant mixture. Reactor efficiency is evaluated by measuring pollutants residual concentrations and CO/CO_2 selectivity as a function of the input energy. The continuous monitoring of CO and CO_2 concentration is realised with respectively an electrochemical analyser and a non-dispersive infra-red analyser.

Experimental results

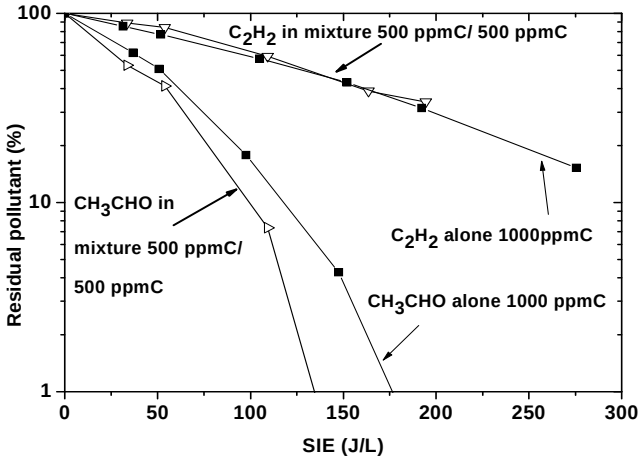

Figure 1 : Comparison of degradation of 1000 ppmC of acetaldehyde and 1000 ppmC of acetylene alone and in mixture in a 94.5% N_2/5%O_2 gas flow

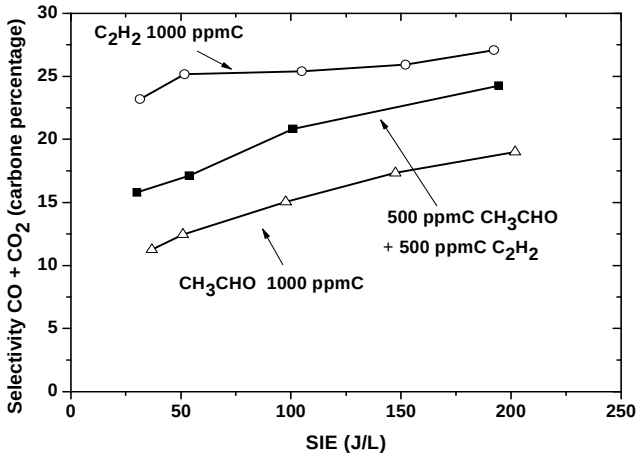

Figure2 : Comparison of global CO and CO_2 selectivity after conversion of pollutants for an initial gas flow containing 1000 ppmC of carbonated species

Experimental results obtained for the removal of an equimolar acetaldehyde/ acetylene mixture are reported on figure 1 and figure 2. It can be firstly observed that in the studied 1000ppmC concentration conditions, the pulse discharge removes pollutants present in the mixture with a good efficiency with 99% of the initial 500 ppmC of acetaldehyde and of 66% of the initial 500 ppmC of acetylene eliminated for a specific input energy inferior to 200 J/L. Besides no real mixture effect can be identified by comparison with mixture containing the pollutant alone (figure 1). However a slight improvement of acetaldehyde removal can be observed in the mixture. Concerning global selectivity in CO and CO_2 (figure 2), values are globally inferior to 30%, meaning a high dispersion of end products distribution, which is not suitable for this type of process.

Modelling part

To improve our knowledge of the chemical phenomena occurring in the discharge, a modelling study has been achieved with a quasi-homogeneous 0D chemical model [3] taking into account 90 species involved in 460 reactions.

This model has been validated by the good accuracy observed when simulated and experimental results for pollutants removal are compared. This good accuracy has allowed us to use this model to compute the consumption flow of the two pollutants in a streamer zone (figure 3 and 4) during one period between the beginning of two successive voltage pulses.

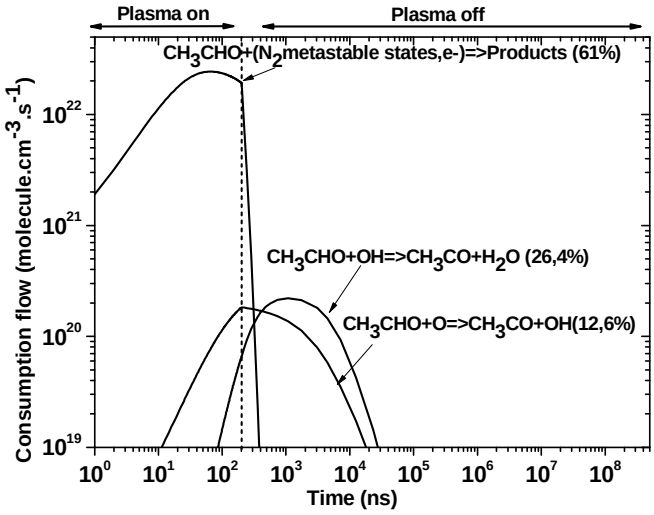

Figure 3 : Computed consumption flow of acetaldehyde for an initial mixture containing 500 ppmC of acetaldehyde and 500 ppmC of acetylene in a 94.5% N_2/5%O_2 gas flow

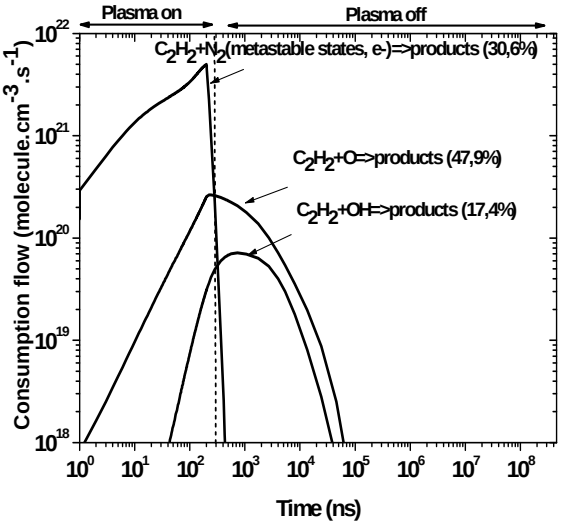

Figure4 : Computed consumption flow of acetylene for an initial mixture containing 500 ppmC of acetaldehyde and 500 ppmC of acetylene in a 94.5% N_2/5%O_2 gas flow

The observation of these consumption profiles have lead us to the conclusion, that, as formerly observed for individual removal of acetylene [3] and acetaldehyde [4] in equivalent experimental conditions, acetaldehyde is mainly decomposed by short life species like N_2 metastable states or electrons when the plasma in on, whereas acetylene is mainly consumed during the post discharge phase by the long life oxygenated species produced during the temporal discharge phase,.

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