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Effect of electric field on the ignition of methane–air mixtures

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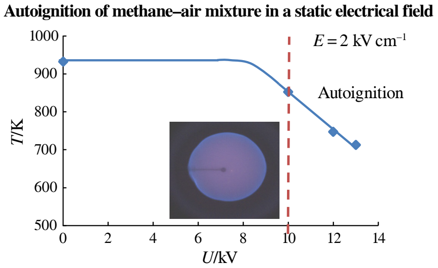
A significant decrease in the ignition temperature of stoichiometric methane–air mixtures in a constant electric field, which did not lead to a breakdown in the gas, was observed experimentally. A kinetic interpretation of this phenomenon is proposed. The discovered effect offers an opportunity of controlling the ignition of combustible mixtures and expanding the temperature and concentration limits of their ignition at the lowest possible expenditure of external energy.



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The ignition,[1,2] propagation,[3,4] and formation of combustion products[5] of fuel–air mixtures determine the performance characteristics of many energy and technological processes. Therefore, the effective control of ignition and its temperature and concentration limits is an important scientific and practical task.[6] Various gas discharges are widely used to control the ignition of combustible mixtures, for example, spark or corona ignition, leading to the formation of local plasma zones.[7] However, spark ignition requires a significant expenditure of external energy,[8] and it does not allow going beyond the concentration limits of flame propagation.

The effect of an electric field on a flame caused by the presence of charged particles as a result of high-temperature chemical ionization[9] has been well known for a long time. On the contrary, the influence of an electric field on the ignition of combustible mixtures has almost not been discussed in detail. It was found that, when a positive voltage of 3.8 kV was applied to a nickel wire located in the center of a reactor, the ignition temperature of mixtures of organic fuels (kerosene, diesel fuel, gasoline, and petroleum ether) with air noticeably increased by about 50–70 °C.[10] The change in polarity led to a decrease in the delay time of autoignition.

A monotonic increase in the upper concentration limit of fuel ignition was observed with an increase in the applied voltage from 0 to 47 kV, both at positive and negative potentials,[11] but a negative potential had a greater effect. The ignition of a CO–O_2 mixture occurred more easily when an electric field was applied to the mixture, and the flame propagation velocity in the longitudinal direct current electric field also increased.[12]

We conducted the first systematic study of the effect of a constant electric field with a voltage of up to 15 kV, which did not lead to breakdown and discharge in gas, on the ignition of stoichiometric methane–air mixtures. A decrease in the autoignition temperature by more than 200 °C was found experimentally. A kinetic interpretation of the phenomenon was proposed.

The experiments were carried out in a static installation described previously.[1,2] A heated spherical-cylindrical stainless steel reactor had a diameter of 120 mm, a total length of 272 mm, and an optical quartz observation window with a diameter of 120 mm and a thickness of 20 mm. Steel balls of various diameters installed in the center of the reactor were used as electrodes (Figure S1, see Online Supplementary Materials).

The minimum temperature of methane autoignition determined under the conditions of these experiments was 660 °C, which is higher than a published value of 580 °C. The difference was probably due to uneven heating of the reactor walls caused by the presence of an optical window for visual observations.

When a negative voltage $U = 10$ kV was applied to the central electrode, the autoignition of a stoichiometric mixture of methane and air occurred already at $T = 580$ °C; that is, this temperature is the lower limit of the autoignition temperature of methane at this voltage. The ignition delay time (IDT) was ~7 ms, which is three orders of magnitude lower than the delay time of autoignition at the minimum autoignition temperature in the absence of an electric field and corresponds to IDT for high temperatures above 1000 °C. At $U = 12$ and 13 kV, the minimum ignition temperature decreased to 475 and 440 °C, respectively (Figure 1). When the polarity of the central electrode was reversed, the mixture did not ignite at temperatures up to 700 °C.

Even a small increase in the reactor pressure, which significantly affects electric discharge processes in gases,[13] increased ignition temperature and IDT. At $T = 520$ °C and $U = 12$ kV, IDT was 7 ms at reactor pressures of 1.0 and 1.1 atm, but it increased to 58 ms at a pressure of 1.2 atm, and the mixture no longer ignited at 1.3 atm.

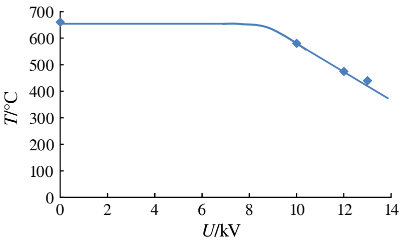

Figure 1 Effect of an electric field on the minimal autoignition temperature of stoichiometric methane–air mixtures at $P = 1$ atm.

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High-speed video recording through a quartz window showed that ignition occurred directly near the surface of the central spherical electrode in the region of a maximum electric field strength; then, a spherically symmetrical combustion wave propagated to the periphery of the reactor (Figure 2). A preliminary estimate of the visible flame propagation velocity allowed us to obtain the normal flame velocity $V_n \approx 120$ cm s^{-1} for a stoichiometric methane–air mixture at $P = 1$ atm, $T = 540$ °C, and the voltage $U = 12$ kV.

An increase in the central electrode diameter from 6 to 7 and then to 13 mm caused a noticeable increase in the ignition temperature in accordance with the expected influence of the electric field strength on this process.

Figure 3 shows the temperature dependence of IDT for a stoichiometric methane–air mixture at a voltage of $U = 12$ kV together with a similar dependence reported previously[2] for the above mixture in the same reactor in the absence of an electric field. In the latter case, an acceptable Arrhenius plot was observed, whereas the value of IDT ≈ 6–7 ms, which is almost three orders of magnitude lower, was found in an electric field. This value of IDT observed in the temperature range 450–580 °C in the electric field corresponds to that observed only at $T > 1000$ °C without an electric field. Up to a lower ignition temperature, this value was almost independent of the reactor temperature, rising sharply near its vicinity. An increase in pressure increased the lower temperature of autoignition and IDT, probably, due to a decrease in the electron free path in the gas.

We proposed a preliminary kinetic interpretation of the results. With a sufficient electric field strength, the constant generation of ions as a result of the interaction of gas molecules with cosmic radiation led to an avalanche-like increase in their number and their achievement of sufficient energy along the free path to excite and ionize gas molecules and the subsequent formation of various radicals. We did not consider the mechanism of formation of active centers in an electric field but assumed that the total rate of generation of radicals $H^\bullet$, $OH^\bullet$, $CH_3^\bullet$, *etc*., in the methane–air mixture was proportional to the field strength and, accordingly, it decreased linearly along the radius of the reactor.

According to reliably verified kinetic mechanisms of methane oxidation,[14] in the studied temperature range of $T < 600$ °C ($T < 900$ K), the generation of radicals led to the formation and accumulation of methyl hydroperoxide CH_3OOH in the system, and its critical concentration reached in the mixture can lead to branched-chain ignition. In accordance with this, the reverse problem was consistently solved: determining the concentration of CH_3OOH necessary for the ignition of the methane–air mixture at a given temperature and realistically permissible IDT of autoignition of 20 s and then determining the required rate ψ_0 of stationary generation of radicals near the central electrode.

A simulation showed that, at a reactor temperature of $T_0 = 475$ °C, it was sufficient to achieve a methyl hydroperoxide concentration of $[CH_3OOH]_{max} = 1$ ppm to ignite the stoichiometric methane–air mixture; thereafter, a gradual rise in temperature was observed during IDT, culminating in ignition with a sharp increase in the concentration of all radicals. For this, a steady-state rate of radical generation $\psi_0 \approx 10^{12}$ molecule cm^{-3} s^{-1} at IDT ≈ 20 s was sufficient, which was achieved due to primary ions (mainly electrons) as a result of their acceleration in an electric field and subsequent interaction with gas molecules. The critical concentration of CH_3OOH required for ignition increased with decreasing initial temperature.

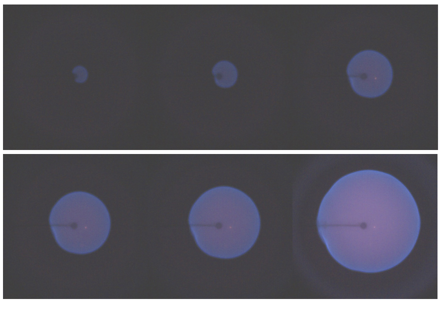

Figure 2 Spherically symmetric ignition wave in a stoichiometric methane–air mixture at $P = 1$ atm, $T = 540$ °C, and $U = 12$ kV.

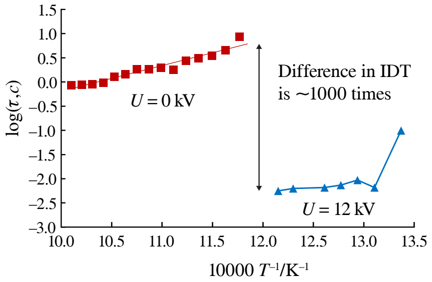

Figure 3 Temperature dependence of IDT for a stoichiometric methane–air mixture at $P = 1$ atm and the voltage $U = 0$ kV (■, published data[2]) and $U = 12$ kV (▲, this work).

Thus, for the first time, we experimentally found a significant decrease in the minimum autoignition temperature of a stoichiometric methane–air mixture in a constant electric field and proposed a kinetic interpretation of this effect.

Online Supplementary Materials

Supplementary data associated with this article can be found in the online version at doi: 10.71267/mencom.7730.

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