Abstract. The influence of pulsed nanosecond silent (or barrier) discharge on the flame velocity was investigated for propane/air mixture in a wide range of equivalence ratios at atmospheric pressure. It was experimentally obtained that blow-off flow rates are more than twice increased as compared to the system without silent discharge. Discharge energy input is less than 1% of burner power. This effect is explained by the formation of atomic oxygen in silent discharge followed by acceleration of limiting chemical reactions.

1. Introduction

The works aimed at applying electric discharge to increase flame velocity (for example, [2]) form a separate group of studies. Faster burning means the possibility to practice combustion of mixtures with low fuel content followed by reducing the formation of NOx oxides. Therefore, the problem of an effective controlling the flame stability is actual nowadays. The goal of the present work is to find mechanisms of flame control.

The easiest way to influence a gas mixture by means of electric discharge is well-known to all motor-car enthusiasts, and this is a spark plug. But in this case spark discharge current is rather large, so mixture ignition is caused by thermal heating of gas. That’s why mechanisms with energy input in flame which is low or insufficient for mixture heating up to spontaneous ignition temperature has special significance.

Flame can be treated as a low-temperature plasma with charged particles energy approximately of 0.1 eV, which mainly consists of molecules and radicals as well as electrons and positively charged ions caused by chemi- and thermo-dissociation. So, besides joule heating, which was mentioned above, there are two main mechanisms which are suggested for the influence of electric field on flame velocity. In weak fields (about couple of hundred volts per centimeter), where field energy is insufficient to excite mixture components, the ionic wind is a determining factor. Under its influence redistribution of charged particles in the combustion zone as well as neutral particles involved in motion by resonance recharging process takes place. It was shown in a classical book of Lawton and Weinberg [3] that such electric forces acting in flame can as more as two hundred times exceed convectional forces. The influence of ionic wind is especially considerable in the case of diffusion flames, where velocities are much lower than in premixed flames [2].

In strong fields processes of chemical kinetics, which are being changed under field influence, play an important role in flame. In this case particles energy should be great enough for electronic and vibrational excitation of molecules, so we mostly imply electrical discharges (corona discharge, streamer discharge and so on).

The method of acceleration of combustion by means of non-equilibrium excitation of gas mixture components allows to change chemical kinetics processes; so it is seemed to be the most promising nowadays. Such excitation can be achieved by using corona discharge. Short (nanosecond) pulses should be used to provide high-performance electronic excitation. Pulsed barrier discharge was selected to avoid the transition of the streamer discharge to the spark form (dielectric barrier limits the maximum current). The form of discharge is non-equilibrium – electron’s temperature is quite high (about 4-5eV), whereas temperature of heavy particles is near to room temperature. Thus there is no any gas heating by the discharge, and energy is put into the vibrational and electronic degrees of freedom of the mixture.

2. Experimental set-up

A set-up consisted of burner, pulse high-voltage power supply, gas supply system and diagnostic system has been used to settle the problem (Fig.1).

The burner body is made of glass. The exit section is a rectangle with 30mm x 2mm dimensions, so flame basis is long enough to work in a 1D geometry. Multipoint brass plate with rectangle section 1mm x 28mm is used as a high-voltage electrode and placed into the nozzle parallel to its walls. Low-voltage electrodes are
made of copper foil, placed into the quartz tubes and positioned near the burner nozzle. The distances between high voltage electrode and quartz tubes can be varied.

One of the advantages of such construction is the fact that voltage and discharge power are limited from above only by high-voltage generator intrinsic properties; the transition from streamer phase to spark form is impossible because of dielectric barrier.

High voltage pulses are produced by pulsed high-voltage generator assembled according to rotating interrupter scheme. Characteristics of the single pulse are the following: amplitude up to 25kV, time of front grow is 10±2 ns, pulse duration on the half-height is 77±5 ns, pulse repetition rate is 1200 Hz. The polarity of pulses could be varied, as well as voltage type: direct or pulse. Pulse’s current and voltage was measured during the experiment; this allowed to obtain the value of energy input.

A blow-off flow rate was chosen as a macroscopic parameter which determines the efficiency of barrier discharge influence on the flame. Blow-off rate is directly connected with rates of chemical processes. Consumptions of mixture’s components were measured to determine blow-off flow rate.

The investigation on active particles in flame was carried out by optical methods using an emission spectroscopy technique.

So, such set-up makes it possible to study burning of premixed gases in a wide range of equivalence ratios (propane-butane mixture used as a fuel and air used as an oxidizer) and generate the non-equilibrium plasma ahead the flame front.

3. Experimental results

It has been found out that influence of the barrier discharge on the flame leads to increase in blow-off flow rate, which is proportional to the discharge power. Nevertheless, the discharge energy input is less then 1% of burner power. Four different cases were studied: direct 25 kV voltage of positive polarity, direct 25 kV voltage of negative polarity, 25kV voltage pulses of positive polarity and 25kV voltage pulses of negative polarity. The comparative analysis of these effects and their influence on blow-off rates is represented on figure 2. It was found out that the pulse 25kV voltage of positive polarity is most effective. It is more efficient than the pulse discharge of negative polarity because cathode-directed streamer is more saturated with active particles than the anode-directed one. On the contrary, negative voltage is more effective in the case of direct voltage discharge (cathode is a good emitter of electrons and corona appears on it). Positive polarity of direct discharge doesn’t provide any significant result.

For pulse voltage, which is the most effective for flame velocity increase, the change of
blow-off flow rate was studied in a wide range of equivalence ratios (0.5-5); see Fig.2. It’s clear that the most strong influence of discharge takes place in the area of \( \phi = 0.65-0.75 \). The change in blow-off flow rate in the system with barrier discharge exceeds 100%.

Pulse voltage monitoring was performed to evaluate the energy inputted into the gas mixture. The energy of one pulse is 8 mJ, and this value corresponds to the mean power approx. of 9 watts. This is less than 1% of chemical power of propane/air mixture burning.

The diagnostic of active particles in flame was fulfilled by means of emission spectroscopy. The results are shown in Fig.3: the barrier discharge spectrum in a propane/air mixture (\( \phi = 1, U = 22kV \)), premixed propane/air flame spectrum (\( \phi = 0.6 \)) and the spectrum of premixed propane/air flame in the presence the barrier discharge (\( \phi = 0.6, U = 22kV \)). Spectra were read within the range of 200-600 nm.

Emission bands in these spectra are identified easily. Bands of \( 2^+ \) system of nitrogen (290-550 nm) are seen well in a barrier discharge. There can be also found \( \gamma \)-bands of NO oxide (220-260 nm) and \( 1^+ \) nitrogen system band (391.4). Bands of \( CH, CN, NO, NH, C_2 \) are existed as well as emission line of \( 0 \rightarrow 0 \) vibrational transition of \( A \Sigma^+ \rightarrow X^3\Sigma^+ \) electronic transition (306.4 nm) in the flame spectrum. When discharge is on, an emission line of \( OH \) 3→2 vibrational transition appears in flame (294.5 nm). The main mechanisms of \( OH \) formation in reaction zone are:

\[
CH + O_2 \rightarrow OH^* + CO \quad (1) \\
H + O \rightarrow OH^*. \quad (2)
\]

OH radical plays an important role in combustion and mainly determines reaction rate as well as normal flame velocity.

Emission profiles of \( C_2 \), \( CH \) and \( OH \) particles are shown in Fig.4. It’s distinctly seen that emission intensity maximum increases, and profile becomes narrower and moves toward the nozzle in the presence of barrier discharge due to the
intensification of the combustion and acceleration of reaction rates. A second emission peak appears near the burner nozzle in the case of OH radical profile due to OH formation in discharge. Such OH behavior confirms importance of OH radical role in the combustion processes. In Fig.5 you can see the OH emission dependence upon the height above burner for different gas consumption in the presence of barrier discharge. Authors assume that barrier discharge influence on flame velocity is the result of excitation of initial gas mixture and formation of active particles before the reaction front. It’s well-known that reagents’ excitation as well as increasing of number of radicals leads to burning acceleration.

In a general case, electric field distribution in a discharge gap is non-uniform and rather difficult for precise description. We can use voltage between electrodes and the gap’s length to get a rough estimate of the reduced electric field. Such estimation gives us the value of 330 Td. Another method of determining reduced field lie in measuring the emission intensity ratio of electronic-vibrational nitrogen transition (detailed description of this technique can be found in [4]). Overestimated value of reduced field obtained by means of this method is equal to 570 Td.

We’ll consider 1D stationary task as an approach. In this case kinetics of plasmochemical process taking place in the pre-combustion zone in the presence of discharge can be divided into three spatially-separated parts: excitation of molecules by electron impact in the barrier discharge, kinetics of excited states and combustion processes. Of course, these areas can intersect each other and it’s worth taking into account state-to-state kinetics as well as the influence of excited particles on the combustion. Unfortunately, the majority of constants of such processes are unknown, that’s why authors state problem to build qualitative model describing burning acceleration by silent discharge.

On the first stage formation of excited particles by the electron impact takes place. Streamer starts from the high-voltage electrode in the presence of high-voltage pulse (about 100 ns). High-energy electrons are generated in streamer head (5-6 eV at the maximum of EEDF). In 5-7 ns streamer overlaps the gap. Only a streamer head (which is quite small) produces excited species as a rule, but the regime exists when high electric field within the total gap interval is reached. This mode takes places after overlapping and lasts until the
spark breakdown. In the case of barrier discharge spark channel can’t appear because of dielectric barrier limits electric current in the circuit. It’s important that in this case field in the gap is a linear function of the length and generation of electrically excited particles occurs along the full length of the interelectrode space quite uniformly. So it’s worth expecting the increase of number of excited species in the gap during the overlap regime because of their additional formation in the streamer channel. The main processes taking place during the electron impact are given below

- Elastic collisions of electrons with molecules;
- Vibrational and electronic excitation of molecules;
- Dissociation;
- Ionization.

The analysis of high-voltage pulse energy branching to the processes given above within the range of our interest (300-600 Td) was performed using a “BolSig” package; results are shown in figure 6. The main part of energy is put into the electronic excitation of molecules (especially N₂) as well as dissociation, and into the nitrogen ionization in the case of strong fields.

After high voltage being turned off, electric field in the gap rapidly decreases and high-voltage electrons in channel disappear. Ionic-molecular reaction practically finishes.

On the second stage that lasts tens of microseconds processes with participation of formed excited particles as well as atomic-molecular reactions take place. Let’s consider the main of them:

- Quenching of electronically excited states of nitrogen molecules. The main channels of their quenching are:

\[ \text{N}_2^* + \text{O}_2 \rightarrow \text{N}_2 + \text{O} + \text{O}. \]  

- Emission of electronically excited singlet states of nitrogen molecules.
- Dissociation of oxygen molecule through its electronically-excited states.

\[ \text{O}_2^* \rightarrow \text{O}^3\Pi + \text{O}^3\Pi \]  

\[ \text{O}_2^* \rightarrow \text{O}^3\Pi + \text{O}^1\Delta \]  

According to energy levels diagram, reaction (5) goes through \( \text{O}_2^3\Sigma^+ \) state, and reaction (4) goes through \( \text{O}_2^A^3\Sigma^+ \) state. The number of O atoms formed in reaction (5) is three times greater than that in (4), so about a half of all atoms are in \( \text{O}^1\Delta \) state. This fact explains appearance of electronically-vibrationally excited OH in the presence of barrier discharge on wavelength which equals 294.5 nm

- Ionization of mixture’s components (N₂ mainly). The main ions – \( \text{N}_2^+ \) and \( \text{O}_2^+ \) – quite quickly transform into the \( \text{O}_2 \) molecule through \( \text{N}_4^+ \) and \( \text{O}_4^+ \) ions
- Translational relaxation of vibrationally excited states of reagents. This is the way the gas heating occurs.

Authors draw a conclusion that at the mentioned times electronic and vibration excitation relaxes into translational movement as well as oxygen dissociation. About 40% of total energy goes to oxygen dissociation. Thus, the third stage can be treated as combustion of new mixture formed at the burner exit as a result of barrier discharge influence.

Calculation gives that concentration of atomic oxygen due to barrier discharge influence is quite high (0.13%); this explains such significant combustion acceleration.

A numerical modeling of combustion and propagation of 1D premixed methane-air flame is performed with the aid of “PREMIX” subprogram of “CHEMKIN” package. This program allows to take into account diffusion of components, thermo diffusion, and heat transfer. The main equations are: the equation of mass conservation, the heat transfer equation, the mass transfer equation and the equation of state. All forward rate coefficients are supposed to satisfy Arrhenius law, and each reaction proceeds according to the law of mass action. Kinetic scheme consists of 17 components and 58 reactions.

The results of modeling are represented on figure 7. Let’s examine modification of the concentration profile of OH radical. It’s distinctly seen that reaction zone becomes narrower with the increase of initial concentration of oxygen atoms. This means that combustion rate is increased too. The main particles taking part in reactions are O,
H, and OH. Oxygen atoms that formed at the burner exit participate in reaction of chain branching. If the temperature is low (there is now gas heating by the discharge), rate of chain breaking is greater than chain branching rate, so initial oxygen concentration isn’t enough to mixture inflammation. Nevertheless, temperature increase happens due to chemical reactions and heat flow from the reaction zone to the nozzle. Temperature rise depends on the atomic oxygen concentration, so flame velocity goes up with oxygen concentration increase.

If concentration of oxygen exceeds some critical (about 0.12%), the model gives an infinity value of flame velocity. This result should be interpreted by the following way. At certain concentration a temperature at which rate of chain branching becomes equal to the rate of chain breaking is achieved due to chemical reaction. The initial oxygen concentration in this case is enough to inflammation, so flame velocity is equal to infinity formally. For every flow rate combustion will be supported. But this situation demands to sustain the critical oxygen concentration at burner exit permanently, and this, in turn, demands infinite discharge power. So in practice critical concentration achieved, than flow rate increases and number of O atoms drops. As a result burning decelerates and, if flow rate is fast enough, can’t sustain temperature gradient required for flame stabilization. The flame velocity increase is determined by the discharge power and is equal to 20% at maximum subcritical oxygen concentration.

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**Conclusions**

The mechanisms of influence of non-equilibrium discharges on the propagation of premixed propane-air flame have been investigated in the present work. Here are the main results of experimental work and numerical simulation:

- Premixed flame acceleration in the presence of different types of discharges has been studied. A comparative analysis of discharges influence on burning rate was performed.
- Changing in premixed flame velocity in the presence of nanosecond pulse silent discharge has been investigated. With energy input less then 1% of burner’s power a combustion of mixtures with fuel content half as great in comparison with system without barrier discharge accomplished. A two times increase in flame velocity has been obtained.
- As a primary mechanism of flame blow-off velocity increase we assume acceleration of limiting reaction followed by excitation of fuel mixture components (i.e. N2) by means of silent discharge.
- A numerical model has been built. The results obtained are in a good agreement with the experimental ones. The combustion acceleration occurs due to the formation of atomix oxygen in front of the reaction zone. It’s well known that excitation of reagents increases the rates of many reactions, so excitation of O2 molecule leads to increase in combustion rate on the whole. Additional heat release in this region and radicals production lead to the blow-off flow rate growth.
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References