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ANALYSIS OF THE IGNITION MECHANISM BY THE COMBINED DISCHARGES

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Successive ignition of hydrocarbons fuels combining an autoignition delay time decrease with a high flame propagation speed is one of the key problems of plasma assisted combustion. The combination of laser and microwave discharges was proposed to use, firstly, to create the fuel-gas mixture ionization before ignition using a laser pulse and, secondly, to ignite the mixture using the microwave discharge as the thermal source. In this paper we present the modeling results of the ignition phenomenon investigations for the case of combined laser-microwave discharge. Ignition delay time as the function of the mixture ionization level formed by a laser pulse and the microwave electric field strength is presented. Ignition mechanism of the combined laser-microwave ignition is analyzed.

KEY WORDS: plasma assisted combustion, ignition, hydrocarbon fuels, laser-microwave discharge.

АНАЛИЗ МЕХАНИЗМА ВОСПЛАМЕНЕНИЯ КОМБИНИРОВАННЫМИ РАЗРЯДАМИ

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Воспламенение углеводородных топлив, сочетающее снижение времени задержки воспламенения с высокой скоростью распространения пламени является одной из ключевых проблем горения, стимулированного плазмой. Для этих целей была предложена комбинация лазерного и микроволнового разрядов, во-первых, для создания области ионизации в топливно-воздушной смеси и во-вторых, для воспламенения смеси микроволновым разрядом, действующим как тепловой источник. В настоящей работе представлены результаты численного исследования процесса воспламенения комбинированным лазерным и микроволновым разрядами. Представлены расчеты времени задержки воспламенения как функции уровня ионизации смеси, формируемого лазерным импульсом, и напряженности электрического поля микроволнового разряда. Проведен анализ механизма воспламенения комбинированными лазерным и микроволновым разрядами.

КЛЮЧЕВЫЕ СЛОВА: стимулированное плазмой горение, воспламенение, углеводородные топлива, лазерный разряд, микроволновой разряд.

АНАЛІЗ МЕХАНІЗМУ ЗАПАЛЕННЯ КОМБІНОВАНИМИ РОЗРЯДАМИ

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Займання вуглеводневих палив, яке поєднує зниження часу затримки запалення з високою швидкістю поширення полум'я є однією з ключових проблем горіння, стимульованого плазмою. Для цих цілей була запропонована комбінація лазерного і мікрохвильового розрядів, по-перше, для створення області іонізації в паливно-повітряній суміші і по-друге, для запалення суміші мікрохвильовим розрядом, що діє як джерело тепла. У даній роботі представлені результати чисельного дослідження процесу займання комбінованими лазерним і мікрохвильовим розрядами. Представлені розрахунки часу затримки запалення як функції рівня іонізації суміші, формованого лазерним імпульсом, і напруженості електричного поля мікрохвильового розряду. Проведено аналіз механізму займання комбінованими лазерним і мікрохвильовим розрядами.

КЛЮЧОВІ СЛОВА: стимульоване плазмою горіння, займання, вуглеводневі палива, лазерний розряд, мікрохвильової розряд.

1. Introduction.

The autoignition delay time for the most of hydrocarbons fuels at atmospheric pressure conditions and elevated temperatures varies in milliseconds range that is unacceptable for the aerospace hypersonic vehicles, operating at high Mach numbers as it leads to a very long ignition region. Therefore any attempts to decrease an ignition delay time and to increase a flame propagation speed are of great interest for applications. Combustion process control by the different kinds of discharges is now a timely topic worldwide mainly due to a possibility of more efficient fuel usage and

reduction of the pollution through ultra-lean combustion.

In recent years the different kinds of discharges were proposed to use as a promoter of combustion process. The nanosecond pulsed discharge in the form of the fast ionization wave as a way of ignition, which allows reducing ignition delay time and increasing the flame velocity due to the production of active radicals is discussed in [1]. The laser discharge as an alternative of a traditional spark discharge is considered as a reliable ignition source, which has applications in the internal combustion engines [2], as well as in the supersonic conditions [3]. The main founding is possibility to organize a multi-point ignition formed by the laser beams, that, in turn, increases the combustion pressure, shortens the combustion and ignition delay time and decreases the induction zone length significantly. The main disadvantages of the laser ignition with the long pulse duration are the complexity of laser beams equipment and the energy loss due to formation of a strong shock wave. As it was shown in [4], the microwave discharge as a thermal ignition source can cause the flame propagation speed enhancement up to 20% at the equivalence ratio 0.5–0.7. The alternative way of combined ignition includes as the laser pulse ability to ionize a mixture, as well as the microwave pulse ability to heat a mixture. As it was demonstrated in [5], [6] rapid gas heating and ignition of methane/air mixtures is achieved with a small amount of femtosecond, non-resonant laser energy and a high power microwave heating pulse. Subcritical microwave fields with pulse energy of <25 mJ have shown strong coupling to the focal volume, containing weak ionization (but negligible heating) due to the sub-mJ femtosecond laser pulse.

2. Mathematical model.

To model the ignition process of methane-air mixture by the combined laser and microwave discharge at atmospheric pressure conditions, the following two stage iteration procedure was used. On the first stage all the rate constants of electron impact reactions such as ionization, dissociation and electronic and vibrational excitation were calculated using the solver S-PLASIG [7] based on the standard electron impact cross sections added by the cross sections data for hydrocarbons from [8]. On the second stage evolution of the microwave discharge plasma parameters is described in the approximation of the plasma uniformity along the axial coordinate, that leads to the approximation of a constant gas density.

The model includes translational and vibrational temperature equations, number density and species concentrations equations. The kinetic model includes the reactions between neutral, excited species, ions and electrons and the reactions caused by an electron impact. The rate constants of methane-air plasma and ethylene-air plasma were taken from [9], [10]. To model methane-air plasma the model incorporates a set of equations for number density of the neutral species (53 neutrals for the GRI 3.0 mechanism [11], 87 neutrals for the C2 mechanism of hydrocarbons combustion [12], positive ions (N_4^+, N_2^+, NO^+, O^+, N^+, O_4^+, $NON2^+$, $NOO2^+$, $O_2N_2^+$, N_3^+, N_2O^+, NO_2^+, O_2^+, $NONO^+$), negative ions (O_2^-, O^-, NO^-, O_3^-, NO_3^-, NO_2^-, O_4^-, N_2O^-, electrons, and excited species $O_2(a^1\Delta)$, $O_2(b^1\Sigma)$, $O_2(c^1\Sigma)$, $O(^1D)$, $O(^3P)$, $N_2(B^3\Pi)$, $N_2(C^3\Pi)$, $N(^2D)$, $N(^2P)$, $N(^4S)$, $N_2(a'^1\Sigma)$ and vibrationally excited nitrogen molecules in the ground electronic state $N_2(x^1\Sigma,\nu)$. The vibrationally excited nitrogen molecules in the ground electronic state $N_2(x^1\Sigma,\nu)$ were treated as single species, i.e. specific vibrational kinetics is not considered, and the microscopic kinetic processes by the levels (V-V exchange) have been neglected. It was assumed, that the vibrational excitation occurred mainly due to the electronically excited species relax to the vibrationally excited nitrogen. All the reactions included in the model are presented in [13].

Equation for the vibrational energy of excited nitrogen molecules ε_V was taken in the form

$$\frac{d\varepsilon_\upsilon}{dt} = \alpha_V Q_E - \frac{\varepsilon_\upsilon - \varepsilon_0(T)}{\tau_{VT}}, \qquad (1)$$

where $\varepsilon_0(T)$ is the equilibrium vibrational energy, α_V is the part of an electromagnetic field energy transferred into the vibrational degrees of freedom, and Q_E is the electromagnetic field energy. Assuming that a deviation from the thermal nonequilibrium is small, for the equilibrium and nonequilibrium vibrational energies Planck's formula was used

$$\varepsilon_0(T) = N\frac{hw_0}{\exp(hw_0/T)-1},$$

$$\varepsilon_\upsilon(T_\upsilon) = N\frac{hw_0}{\exp(hw_0/T_\upsilon)-1},$$

where hw_0=0.28 eV is the energy of the first vibrational level of nitrogen molecules.

For the vibrational-translational relaxation time τ_{VT} the expression proposed in [14] was used, which includes the correction, depending on the oxygen atoms concentrations:

$$\frac{1}{\tau_{VT}} = 7.0\cdot10^{-10} N\cdot\exp\left(-\frac{141}{T^{1/3}}\right) + \alpha_o\cdot5.0\cdot10^{-12}\cdot N\cdot\exp\left(-128\cdot T^{-1/2}\right),$$

where N is the number density of mixture, α_o is the mole fraction of the atomic oxygen.

It is known that the use of the equation for ε_υ based on the Landau-Teller formalism gives satisfactory results until $T_m \sim hw_0/k_B$, at temperatures higher than T_m anharmonicity effects can influence on the vibrational temperature distribution. To take into account the possible vibrational anharmonicity the phenomenological correction of τ_{VT} proposed in [15] was used in the form

$$\tau^{*}_{VT} = \tau_{VT} \cdot \left[\frac{1-\gamma \exp\left(-T_m / T_\upsilon\right)}{1-\exp\left(-T_m / T_\upsilon\right)} \right]^2,$$

where γ is the adiabatic index.

Methane influence on the vibrational relaxation of nitrogen was neglected. According to the experimental measurements of the acoustic attenuation in the mixture of nitrogen and methane at P=1 atm, T=300 K in [16], dependence of the acoustic attenuation on the methane concentration is weak except very high frequencies of 10^5 –10^6 Hz. Assuming that the acoustic attenuation mainly results from the molecular relaxation processes the assumption of a negligible influence of a methane content on the nitrogen vibrational relaxation was accepted. Influence of the vibrational nonequilibrium on the mixture heat capacity was accounted by the specific heat correction in the form

$$C_p = C_p^{*} - \frac{N_{N_2}}{\rho} \frac{d\varepsilon_V}{dT}$$

where N_{N_2} is the molar fraction of nitrogen molecules, $C_p^{*} = \frac{1}{N} \sum_{i=1}^{n} C_{pi} n_i$ is the mixture specific heat.

The part of an electromagnetic field energy transferred into the vibrational excitation of the nitrogen molecules was taken as a function of the reduced electric field by interpolation of the data presented in [17]. For air-fuel mixture we corrected the energy division on the percentage of nitrogen in the mixture. Influence of the vibrational temperature on the reaction rate with the participation of neutral molecules was neglected, but possibility of the vibrational excitation influence on the dissociation, detachment and excitation reactions was taken into account by multiplying the corresponding reaction rates by an enhancement factor $k_V = 10^{Cz/\theta^2}$ as in [18], where C=28.3, $z = \exp(-h w_0 / k_B T_\upsilon)$, $\theta = E/N$ in 10^{-16} V·cm^2.

The approximate electric field profile of the microwave heating pulse was chosen in the form of a rectangular pulse with the maximum electric field value $E_{max} = \beta \cdot E_b$, where $\beta < 1$ is the problem parameter, $E_b = 120$ Td is the breakdown electric field value at atmospheric pressure conditions. Time of the energy input on this stage was varied with the step extension modeled by the time of the microwave energy input t_m.

Question of the initial conditions for the microwave heating pulse after the femtosecond laser pulse is still open. The only thing which is quite clear is that the concentration of negative ions formed after the laser pulse is low and can be neglected. Oxygen molecule with the potential U_i=12.1 eV and methane with the potential 12.98 are the species which are easier to ionize compared with nitrogen with the ionization potential U_i=15.6 eV, therefore predominance of O_2 molecules ionization in the total electron density is expected. This statement well correlates with the numerical data of the calculated ionization rates of nitrogen and oxygen [22], which are practically linear functions of the laser intensity and differ in orders of magnitude. At the same time in [19] the authors presented the experimental data concerning propagation of fs Ti: sapphire laser pulses in air and detected the photoemission of N_2^+ ions jointly with the excited states on N_2 molecules $N_2(B^3\Pi)$, $N_2(C^3\Pi)$, formed through dynamic multiphoton absorption in the laser field. Formation of the excited states of nitrogen by a laser fs pulse is confirmed by the recent measurements of the flame speed of a laminar premixed methane-oxygen/nitrogen flame presented in [20]. For the pure methane as it was shown in [21] the process of ions formation by the femtosecond laser strictly depends on the laser pulse duration and the main positive ions formed are CH_4^+ with small amounts of CH_2^+ and H^+ but an increase of the femtosecond pulse duration causes appearance of the double-charged ions. A brief review of the ultrashort laser pulse filamentation processes in various transparent media is presented in [22] where authors mentioned that for a filament laser intensity of 5×10^{13} W/cm^2 the electrons densities achieved in air at 800 nm reaches 10^{16}–10^{17} cm^{-3}. It gives us an estimation of the mixture ionization level after fs laser pulse about 10^{-2}–10^{-3}.

As a way of the indirect evaluation of the initial conditions for the microwave heating pulse we were oriented on the experimental data presented in [10] where the successful ignition moment was indicated in a lean methane-air mixture ignited by the combined laser-microwave discharges. According to the presented data of a flame kernel formation at different moments the ignition delay time varies in limits $5\ \mu s \le t < 50\ \mu s$. Taking into account that in the lean methane-air mixture methane content is low and neglecting by the double charged ions formation due to a short fs laser pulse duration it was assumed that the main positive ion formed after fs laser pulse could be O_2^+ and N_2^+.

3. Simulation results and discussion.

To understand the ignition mechanism in the case of the combined laser –microwave ignition two important questions need to be answered. What level of the mixture ionization and the microwave filed strength allows a lean mixture to be ignited with the ignition delay time in a microsecond range, and what is a role of ions and excited molecules in this process? What level of the vibrational nonequilibrium can be created by the microwave heating pulse and how it influences on the ignition process? To answer these questions we consider the case of the lean methane-air mixture ignition (equivalence ratio $\varphi = 0.3 - 0.8$). Initial conditions for the microwave heating pulse with duration of $t_{MW} = 0.5 \div 2\ \mu s$ were chosen as the ionization fractions of O_2^+ and N_2^+ with the ionization level of 10^{-3}.

The moment of ignition was indicated as a maximum on the heat release rate dependence on time after the microwave discharge action. This maximum is very close to the peaks on the HCO, CH and OH dependence on time after the discharge action (within

2–3%). All these quantities were used as an indicator of the fuel-air mixture ignition moment.

The calculated dependencies of the translational and vibrational temperatures as functions of the microwave electric field strength are presented in Fig. 1, 2.

It is seen, that at ionization level of $\alpha = 10^{-3}$ for al the values of the microwave field strength during the microwave heating pulse, except very high values $E_{MW} \geq 0.7 \cdot E_b$, the strong translational-vibrational nonequilibrium is observed. We consider vibrational mode parameter $\xi = \frac{T_\upsilon - T}{T}$ at the end of a microwave heating pulse as a main indicator of the level of nonequilibrium. It was obtained that depending on the problem parameter β the calculated vibrational temperature is twice (at $\xi \sim 1$ and $\beta = 0.55$) or three times (at $\xi \sim 2$ and $\beta = 0.6$) of translational temperature for the equivalence ratios of $\varphi = 0.3 - 0.8$. Note that this tendency observed for the different equivalence ratios is a consequence of the input microwave power division, as the most part of microwave energy goes to the vibrational excitation in the range of the considered microwave field strength of 50–70% of breakdown.

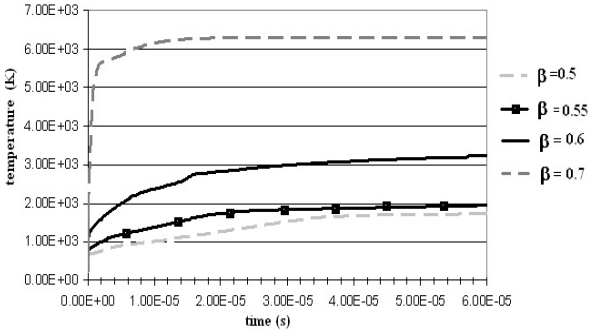

Fig.1. Time dependence of the translational temperature as a function of the parameter β at $\alpha = 10^{-3}$ for methane-air mixture, $\varphi = 0.8$, $t_{MW} = 2\ \mu s$, $(E_{MW} = \beta \cdot E_b)$.

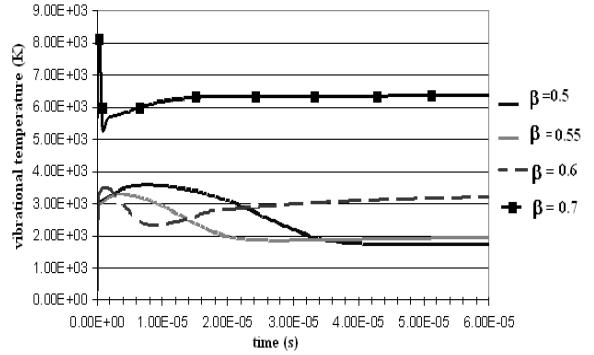

Fig.2. Time dependence of the vibrational temperature as a function of the parameter β at $\alpha = 10^{-3}$ for methane-air mixture, $\varphi = 0.8$, $t_{MW} = 2\ \mu s$.

It should be noted that the numerical calculations have shown that ignition can occur at very low equivalence ratio of the methane-air mixture $\varphi = 0.3$ if the microwave field strength $E_{MVW} > 0.5 \cdot E_b$ and the mixture ionization level is of $\alpha \geq 10^{-3}$. It was also obtained that for the all cases of the successful ignition for the methane-air mixture the ignition delay time is changed in a microsecond range if the vibrational mode parameter $\xi \sim 1.1$, and with the parameter ξ shifts to $\xi \sim 2$ the ignition delay time t_d is changed to the values of tens microseconds (Fig.3).

For the case of the high microwave field strength at $\beta = 0.7$, when the V-T relaxation occur during the microwave heating pulse the parameter ξ=0.0, and the calculated values of t_d is lower than microsecond. The calculations results have shown that the role of the microwave heating pulse in ignition process for a case of the combined laser- microwave ignition is not only to heat the mixture till the autoignition temperature but also to produce a sufficient number of the combustion process precursors due to the electron impact reactions.

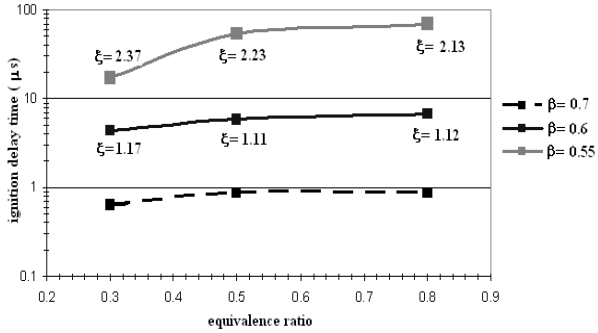

Fig.3. The ignition delay time for the case of the combined laser-microwave ignition at $\alpha = 10^{-3}$, methane-air mixture, $t_{MW} = 2\ \mu s$.

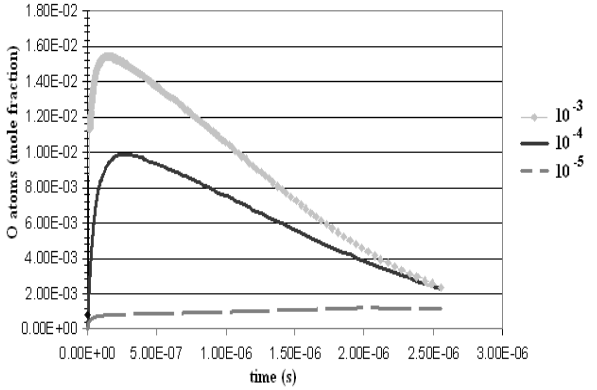

Fig.4. The oxygen atoms dependence on time for different initial ionization level of the methane-air mixture at $\varphi = 0.5$, $\beta = 0.55$, $t_{MW} = 2\ \mu s$.

Note that influence of the ionization, detachment and attachment rates enhancement by a vibrational excitation is weak as the enhancement factor $k_V \sim 1$ in the conditions of the problem. The following scenario of an ignition is proposed. On the first stage, oxygen atoms are formed due to the electron impact reactions such as $O_2 + e^- \rightarrow 2O + e^-$ and $O_2^+ + e^- \rightarrow 2O$, than the main

path of O atoms production is changed to the well-known two stages mechanism [23] with the participation of the excited nitrogen molecules in $N_2(B^3\Pi)$, $N_2(C^3\Pi)$ states. It gives the maximum mole fraction of oxygen about $n_o = 10^{-2} \div 10^{-3}$ for the mixture ionization level of $\alpha = 10^{-4} \div 10^{-3}$ (Fig.4). It is seen from Fig.4 that the oxygen atoms production rates during the microwave heating pulse depend on the initial ionization level created by a seed laser pulse, and at the low values of α ($\alpha = 10^{-5}$) ignition do not occur even at the higher microwave field strength. With the microwave field strength of 50% of breakdown ($\beta = 0.5$) the mixture ignition was obtained only for the methane-air mixture with the equivalence ratio $\varphi = 0.8$. For $\varphi = 0.5$ and $\varphi = 0.3$ for the same $\beta = 0.5$ the maximum translational temperature does not exceed 500 K that does not result in a successful ignition, the similar situation is realized for all the considered equivalence ratios at $\beta < 0.5$. At the same tine an increase of the time of the microwave energy input above the definite value does not lead to an automatic decrease of the ignition delay time (Fig.5). In this case we deal with so called saturation effect as for the spark ignition.

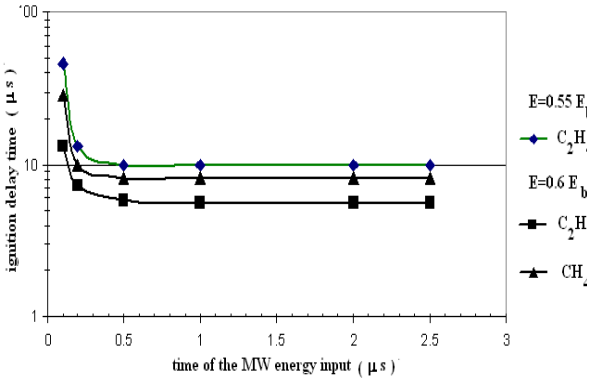

Fig.5. An ignition delay time dependence on the microwave energy input time for the lean mixtures of methane and ethylene ($\varphi = 0.5$).

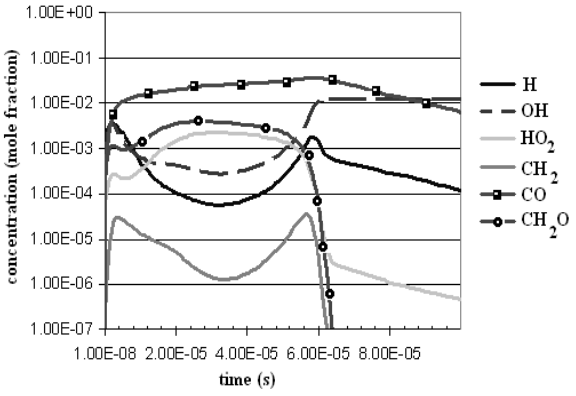

Fig.6. The mixture components dependencies on time for methane-air mixture at $\varphi = 0.5$, $\beta = 0.55$, $t_{MW} = 2\ \mu s$.

The selected mixture components dependence on time is presented in Fig.6 for the case of the lean methane-air mixture of $\varphi = 0.5$ at $\beta = 0.55$ and $t_{MW} = 2\ \mu s$. The moment of ignition is clearly indicated by the peak on the radical OH dependence on time, the key pathway of OH formation in this case is governed by the reaction with HO_2.

Conclusions. We present results of the kinetic modeling of the combined laser and microwave discharge. The main mechanism of ignition in this case is realized as follows. The laser fs pulse governs the branching chain reactions creating the minimal electron and positive ions density needed for the initiation of the electron impact reactions. The microwave heating pulse heats the mixture and additionally produces a sufficient number of the oxygen atoms and OH radicals to initiate ignition. The ignition delay time dependence on the level of the vibrational nonequilibrium created by the microwave pulse was obtained. The calculated ignition delay times for the case of interest vary in limits of microseconds to ten microseconds that well correlates with the experimental data.

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