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Interaction of the laminar flames of methane–air mixtures with close-meshed spherical and planar obstacles in a closed cylindrical reactor under spark discharge initiation

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The spark initiated flames of dilute stoichiometric natural gas–oxygen mixtures in close-meshed aluminum spheres with a mesh size of 0.1–0.2 mm^2 do not propagate through the spheres but always propagate through planar meshed obstacles of the same mesh size. It is found that the features of flame propagation under simultaneous initiation at the opposite butt-ends of a cylindrical reactor differ markedly from those under initiation from a single discharge.

The influence of obstacles located in volumes filled with combustible mixtures on the propagation of a flame front (FF) has been investigated for a long time.[1,2] It is known that, if the composition of a gas mixture is far from ignition limits, the FF velocity in the presence of obstacles can grow to supersonic values.[3,4] The most prominent aspect in the investigation of these accelerated flames is caused by explosion safety problems.[5] The influence of obstacles[1] can be manifested doubly: in the maintenance of a supersonic wave and in the quenching of a supersonic wave as a result of heat losses.

It is possible to reduce the above problem to an initial stage of laminar flames propagation.[6] The interaction of FF with obstacles results in the development of FF instability and acceleration. However, the contact of FF with obstacle surface leads to an increase in the contribution of heterogeneous reactions, in particular, chain termination,[7] which causes flame suppression. It was shown[8] that spark initiated flames of lean hydrogen–air mixtures propagated through meshed aluminum spherical obstacles with a mesh size of 0.04–0.1 mm^2; the flame of 15% H_2 in air after obstacle was accelerated; acoustic gas perturbations occurred in the reactor; the smaller diameter of the spherical mesh caused the earlier start of the acoustic perturbations. However, the FF of a stoichiometric natural gas (NG)–air mixture was not accelerated after obstacle; acoustic perturbations were missing. The conclusion was made that the active centers of methane and hydrogen combustion, which determine flame propagation, have different chemical nature.

Thus,[8] chain termination contributes significantly to the interaction of FF with obstacles in the case of NG–air mixtures. Therefore, it is of practical interest to investigate NG combustion in the presence of meshed obstacles of different shape under different flame initiation conditions in a larger reactor. The aim is to estimate the effectiveness of close meshed obstacles for methane flames suppression.

The experiments were performed with the dilute stoichiometric mixtures of NG with oxygen, CO_2 and Kr at initial pressures of 100–200 Torr and 298 K in the pumped out horizontally located cylindrical quartz reactor 70 cm in length and 14 cm in diameter. The reactor was fixed in two stainless steel gateways at butt-ends (Figure 1), supplied with inlets for gas pumping and blousing and a safety shutter, which swung outward when the total pressure in the reactor exceeded 1 atm. Two pairs of spark ignition electrodes were located near the opposite butt-ends of the reactor; the pairs were connected to power supply consistently to provide two simultanious spark discharges. Each pair could be short circuited to provide only one discharge. The spherical mesh was fixed on one pair of electrodes if necessary, as shown in Figure 1. This spherical mesh consisted of two hemispheres fastened by a spring. Thus, the volume included in meshed sphere and external reactor volume contacted only through the mesh of aluminum wire. Meshed spheres 8 cm in diameter (wire diameter of 0.3 mm, cell size of 0.1 mm^2), d = 10 cm (wire diameter of 0.35 mm, cell size of 0.15 mm^2), d = 13 cm (wire diameter of 0.5 mm, cell size of 0.25 mm^2) were used. The surface of aluminum is always covered with its oxide. Hence, the meshed surface consisted of aluminum oxide, which effectively breaks reactionary chains.[6,8] In other experiments, one of planar meshed Al obstacles 14 cm in diameter (either with a wire diameter of 0.3 mm, cell size of 0.1 mm^2; or with a wire diameter of 0.5 mm and a cell size of 0.25 mm^2) was placed in the reactor at 1/4 or 1/2 of its length.

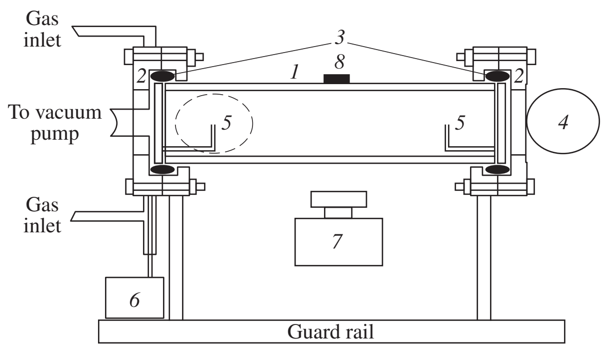

Figure 1 Experimental installation: (*1*) quartz reactor, (*2*) stainless steel gateway, (*3*) silicone laying, (*4*) stainless steel shutter, (*5*) spark electrodes with meshed sphere d =13 cm, (*6*) power supply, (*7*) Casio Exilim F1 Pro high-speed color movie camera, and (*8*) Ritmix microphone.

The combustible mixture (15.4% NG + 30.8% O_2 + 46% CO_2 + + 7.8% Kr) was prepared, and CO_2 was added to decrease FF velocity and to enhance the quality of filming; Kr was added to diminish the discharge threshold. The reactor was filled with the mixture to a necessary pressure. Then, spark initiation was performed (the discharge energy at each pair of electrodes was

1.5 J). Speed filming of ignition dynamics and FF propagation was carried out from the side of the reactor (Figure 1) with a Casio Exilim F1 Pro color high-speed digital camera (frames frequency of 60–1200 s^{-1}).[9] The filming was turned on at arbitrary moment before initiation. The video file was stored in computer memory and its time-lapse processing was performed.[10] NG contained 98.8% methane and 1.2% propane, and other reagents were chemically pure.

The pressure change in the course of combustion was recorded by a piezoelectric gage synchronized with the discharge. Acoustic perturbations were recorded with a Ritmix sensitive microphone (up to 40 kHz). In the course of each experiment, the level of external noises was minimized. The audio recording was turned on at arbitrary moment before initiation. The audio file was stored in computer memory and analyzed with the Spectra Plus 5.0 software package.

In all experiments, the flame initiated in spherical meshes did not propagate through the cells of aluminum grid [Figure 2(*a*)] in agreement with published data[8] denoting that at atmospheric pressure at inner surface of the spherical mesh FF practically stops. Figure 2(*a*) shows that, under our conditions (total pressure of 160 Torr), combustion is observed only inside the spherical mesh for several seconds at the expense of diffusion of unreacted gas into the sphere. The disappearance of NG–air flame outside of the meshed obstacle has no explanation in terms of the thermal theory.[6] One can assume that the attenuation of NG–air flame is connected with the intense heterogeneous termination of active intermediate products of combustion on the surface of the spherical mesh (Al_2O_3); this is the possible reason of the zero velocity of NG–air flame in the vicinity of a meshed obstacle. Thus, the stable intermediate products of combustion (for example, hydroperoxides) diffusing through aluminum grid cells can again initiate flame propagation outside of meshed sphere if the gas mixture outside of the sphere is flammable (*e.g.*, at 1 atm[8]).

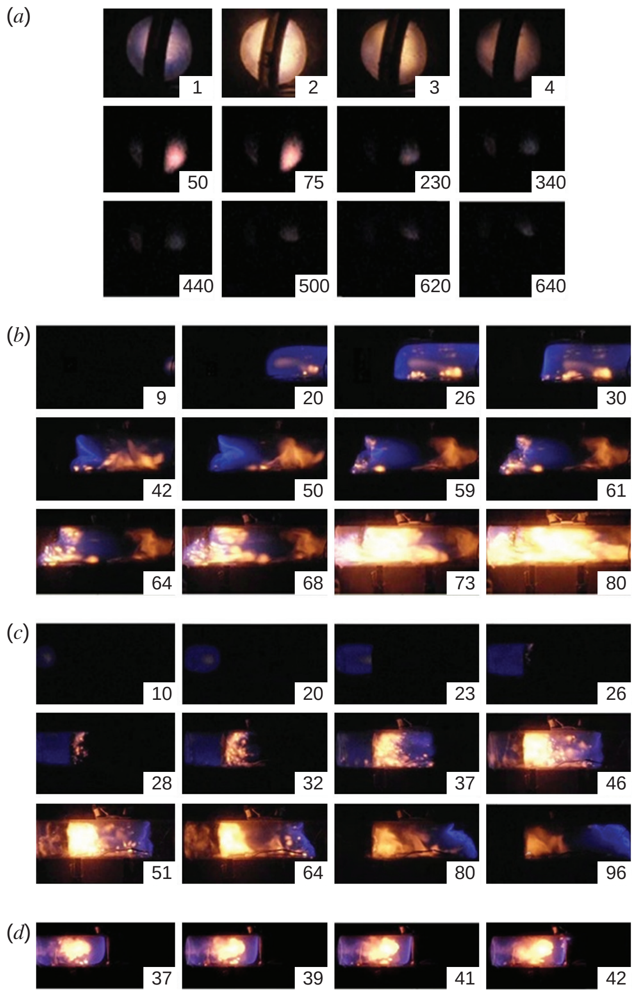

Figure 2 High-speed filming of FF propagation in the gas mixture at an initial pressure of 160 Torr. (*a*) In the spherical mesh, *d* = 13 cm, single spark discharge (1.5 J) in the sphere, speed of filming 60 frames s^{-1}; (*b*) in quartz reactor in the presence of spherical mesh, *d* = 13 cm, double spark discharge (1.5 J) as is shown in Figure 1, speed of filming 600 frames s^{-1}; (*c*) in quartz reactor in the presence of planar meshed obstacle (diameter of wire 0.5 mm, cell size 0.25 mm^2) placed at the 1/4 of reactor length, single spark discharge (1.5 J) on the left, speed of filming 600 frames s^{-1}; (*d*) in quartz reactor in the presence of planar meshed obstacle (diameter of wire 0.5 mm, cell size 0.25 mm^2) placed at the 1/2 of reactor length, single spark discharge (1.5 J) on the left, speed of filming 600 frames s^{-1}. The figure on a frame corresponds to frame number after discharge.

In this work, two similtaneous discharges inside and outside the spherical mesh were carried out [Figure 2(*b*)]. After initiation of FF with outer discharge blue FF propagates from right to left and reaches the spherical mesh [frames 9–61, Figure 2(*b*)]; then, the secondary yellow FF occurs and propagates in an opposite direction from left to right [frames 64–80, Figure 2(*b*)]. Note that the combustion in the spherical mesh placed on the right electrode also takes place [Figure 2(*a*)], but the camera is not sensitive enough to record slow flame in the sphere due to higher speed of filming (600 frames s^{-1}) in Figure 2(*b*) as compared with Figure 2(*a*). It means that chemical transformation in blue 'cool'[11] FF (frames 9–61) propagating to the left is incomplete and some active products of NG combustion in the spherical mesh can initiate the secondary yellow 'hot'[11] FF propagating in opposite direction over incompletely reacted mixture chemically and thermally activated with primary blue FF (frames 64–80). It is in compliance with the results of our earlier work,[12] where we showed that the color of FF in a hydrocarbon–air mixture in a heated cylindrical reactor is always yellow ['hot' flame, Figure 2(*b*)[12]], though FF at initial room temperature in the same mixture and in the same reactor is blue ['cool' flame, Figure 2(*a*),(*b*)[9]]. Notice that the color of blue methane flame is mainly due to CH (431 nm) and CH_2O (470 nm) luminescence, and the yellow color of flame is caused by the emission of Na atoms excited in 'hot' flame and soot particles emission.[11]

Note that the incompleteness of NG conversion during flame propagation from an initiating source could make a contribution both to the origination of so-called Mache effect, which is known to reduce explosion pressure below the one for uniform temperature distribution[11,13] (because gas volumes during combustion have different initial conditions and temperature[6]), and to a deflagration-to-detonation transition (DDT).

Under our conditions, FF in a dilute stoichiometric NG–oxygen mixture never propagates through meshed spheres but always propagates through planar close-meshed obstacles [Figure 2(*c*),(*d*)], although Figure 2(*c*) (frames 23–28) shows that the velocity of FF near the planar obstacle considerably decreases. Seemingly, FF should interact at the same time with the whole obstacle surface to be suppressed, *i.e.*, the curvature of FF should correspond to the curvature of an obstacle as in the case of the spherical mesh. However, as can be seen in Figure 2(*d*), the almost planar FF formed at the reactor center also propagates through the planar obstacle placed at 1/2 of the reactor length. Probably, the observed influence of obstacle shape is connected with the structure of fluxes arising at the propagation of originally spherical FF in the cylinder.

As can be seen in Figure 2(*a*),(*b*), the features of flame propagation at single initiation and simultaneous double initiation from the opposite butt-ends of the reactor differ markedly. It means that investigation into propagation of flame fronts caused by two or more initiating impulses is of specific interest especially for the problems of explosion safety, *e.g.*, fires extinguishing when several sources of flame initiation can occur.

We studied the propagation of counterflames by simultaneous initiation at opposite butt-ends of the reactor, in particular, with the excitation of acoustic perturbations during combustion of the gas mixture. The results of high-speed filming of FF propagation in the gas mixture initiated with two opposite simultaneous discharges and single discharge at an initial pressure of 160 Torr in a quartz reactor in the presence of a planar meshed obstacle placed at the 1/4 of reactor length are presented in Figure S1(*a*) (see Online Supplementary Materials) and Figure 2(*c*), respectively. The FF also propagates through a planar obstacle in the presence of counterflame. The dependences of change in total pressure on time for flame propagation under specified conditions are shown in Figures S1(*c*) and 3(*b*), respectively. As is seen, the change in pressure (proportional to warming-up or heat release per unit time[6]) under initiation with two opposite discharges is more than twice as large as under single discharge. In addition, the propagation of counterflames is always accompanied by characteristic sharp sound; in this case, acoustic perturbations of gas can be recorded [Figure S1(*b*)] and the shutter swings outward. Note that flame propagation under conditions of single initiation is not accompanied by any noticeable sound effect, the shutter does not swing, only weak acoustic perturbations directly relevant to the flame propagation are observed. Figures S1(*b*) and 3(*a*) show the time dependences of acoustic amplitude for flame propagation under conditions of single initiation. Acoustic perturbations [~1500 Hz, Figure S1(*b*),(*d*)], being about ten times more intense in the case of initiation at opposite reactor butt-ends, were observed before shutter swung (38.9 s). Then, the characteristic vibrations of the cylinder with one end open were observed (141 and 281 Hz). Notice that calculated values for characteristic vibrations for our reactor divided with planar obstacle at 1/4 of its length with one end open according to published equations[14] are 145 and 285 Hz.

The results directly indicate that counterflames propagation in a cylindrical reactor caused by simultaneous double initiation at opposite butt-ends occurs with greater warming-up and more intense acoustic perturbations, as compared with flame propagation from a single initiating source.

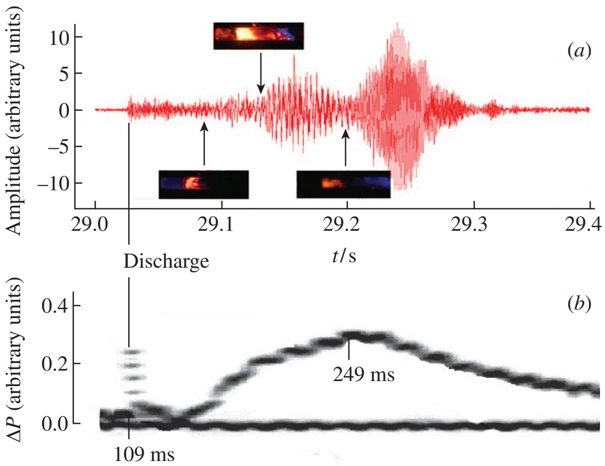

Figure 3 (*a*) The time dependence of acoustic perturbations amplitude in FF propagation in the gas mixture at an initial pressure of 160 Torr in a quartz reactor in the presence of planar meshed obstacle placed at the 1/4 of reactor length, single spark discharge (1.5 J) on the left [see Figure 2(*c*)], speed of filming 600 frames s^{-1}. Several corresponding frames from Figure S1(*a*) are shown. (*b*) Oscillogram of the time dependence of pressure change.

Thus, the spark initiated flames of dilute stoichiometric NG–oxygen mixtures in close-meshed aluminum spherical obstacles with a mesh size of 0.1–0.2 mm^2 do not propagate through the mesh but always propagate through planar meshed obstacles of the same mesh size. Double spark initiation (in the spherical obstacle and outside of it) leads to the occurrence of secondary hot flame front propagating over the whole reaction volume. It indicates the existence of two stages of the kinetic mechanism of NG combustion. The first stage corresponds to the propagation of a ‘cool’ blue front of incomplete chemical conversion; the second stage is due to the fast chemical transformation of products of the incomplete oxidation of NG. The flame propagation at simultaneous double initiation at opposite butt-ends of the reactor differs markedly from that at single initiation. It argues that studies on the propagation of flame fronts caused by two or more initiating impulses are essential for the problems of explosion safety, when several initiation sources can occur.

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Online Supplementary Materials

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.mencom.2013.05.015.

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