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Shock-Tube Ignition Study of Methane in Air and Recirculating Gases Mixtures

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To investigate the effect of recirculated combustion products on the combustion process of a typical gas-turbine engine, shock-tube and modeling investigation of ignition delay time was performed. Different mixtures of CH_4-O_2-Ar-N_2–H_2O were tested. Results showed that replacing N_2 by combustion product components, CO_2 and H_2O, at concentrations and temperature range, which are typical for the flameless oxidation regime, did not affect the ignition delay time. The temperature range of the shock-heated samples was 1350–1800 K, and the pressure range was 5–10 atm. Modeling calculations were carried out with two mechanisms, and they both showed similar tendency. The correlation between the experimental data and calculations is discussed.

I. Introduction

THE design and development of low-emissions aero and industrial gas-turbine combustors is very challenging because it entails many compromises. Satisfying the projected CO and NO_x emissions regulations without relaxing the conflicting requirements of combustion stability, efficiency, pattern factor, and partial engine loading capability is a significant but difficult task. Advanced low-emission combustor design are described by Lyons and Niedzwiecki,[1] Gupta,[2] Lefebvre,[3] Moore,[4] Wulff and Hournouziadis,[5] Correa,[6] and Cavaliere and Joannon.[7] Some combustor types have been investigated and implemented; others are only at an early development stage. One of the methods for NO_x emission reduction that was intensively studied and applied is "flue-gas recirculation." This method is effective in lowering the peak temperatures and thereby the formation of thermal NO_x.

The effect is achieved by the mixing of hot combustion products with fresh air. It increases the inert components of the mixture, reduces the oxygen concentration, and thus lowers the reaction/combustion temperature. The practical feasibility of this method has been investigated by Wilks and Gerhold,[8] who found significant reduction of NO_x (50%) with recirculation rates of about 20%. Recirculation rate definition used in this work is different from the one used in other studies (for example, see Wunning and Wunning[9]). But the flue-gas recirculation method is limited by the maximal quantity of recirculated gas because in normal ambient air vitiation can lead rapidly to blowoff.

Figure 1 shows the relations of CO_2, H_2O, and O_2 content found in different practical installations with recirculation of combustion products. This process was intensively investigated during recent years (see Refs. 9–12). The combined effect of high air temperature and increased content of inert components in the reaction zone on the flame stability was studied by Katsuki and Hasegawa.[13] They found that the temperature of the inlet gases has to be increased if the oxygen concentration is to be lowered. Actually, oxygen content as low as 3% can sustain combustion when its reaction zone is preheated up to 1200 K.

Efforts to apply flue-gas recirculation to industrial gas turbine and jet engines was made by Tabacco et al.,[14] where they investigated trends of flameless oxidation regime in gas turbine. Levy and Arfi[15] proposed a new design of the adiabatic combustor for industrial gas turbine and aircraft jet engines.[16] Thermodynamic and geometric aspects of this combustor were described by Levy and Arfi[15] and Levy et al.[17]

The aim of this work is to study the influence of flue-gas addition to combustion air on the ignition delay time, which has a significant effect on combustion stability, and to compare the results with engineering results in engines and search for incompatibilities. The study was performed in a shock tube, where the experiments were set at conditions as close as possible to recirculating engine data, and compared with modeling predictions.

II. Ignition Delay Times Measured in Shock Tubes, State of the Art

The ignition delay time measured in a shock tube is the time elapsed between the rapid (about a microsecond) temperature rise of the system caused by the shock arrival and the onset of temperature caused by the ignition. This is the time during which radicals are formed and participate in reactions involving fuel and oxygen. (Nitrogen, because of its high bond energy, dissociates and forms radicals only at temperatures beyond 2000 K.) Radicals are accumulated as a result of chemical reactions/collisions until they achieve a threshold concentration where ignition/explosion occurs.

The ignition delay time in a well-stirred reactor, as measured in a shock tube, was summarized in many papers.[18–26,**] A detailed paper about methane oxidation and ignition was written by Spadacini and Colket.[27] It was shown by Lifshitz[18,19] and Lifshitz et al.[20] that the ignition delay time obeys an Arrhenius-like rate equation, which was erroneously referred to Semenov:

$$\tau = 10^x \exp(+E/RT)[\mathrm{Fuel}]^\alpha[\mathrm{O_2}]^\beta[\mathrm{Diluent}]^\gamma \mathrm{s}$$

where [ ] are the reactant concentrations, α, β and γ being the power dependencies of these concentrations, R is the universal gas constant, and T is the static temperature of the mixture in K.

The ignition delay times of methane-oxygen mixtures diluted in argon were shown[20,21] to fit the Lifshitz equation

$$\tau = 10^{-13.44} \exp(+46.5/RT)[\mathrm{CH_4}]^{0.33}[\mathrm{O_2}]^{-1.03}[\mathrm{Ar}]^0 \mathrm{s}$$

It was shown by Grillo and Slack[24] that methane-oxygen mixtures diluted in nitrogen show the same ignition delay as methane oxygen diluted with argon, but this fact was fully investigated by Peterson et al.[25,26] who have shown that there is absolutely no difference if the

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** Data available online at http://www.me.berkeley.edu/gri_mech/ or ftp://unix.sri.com/gri/.

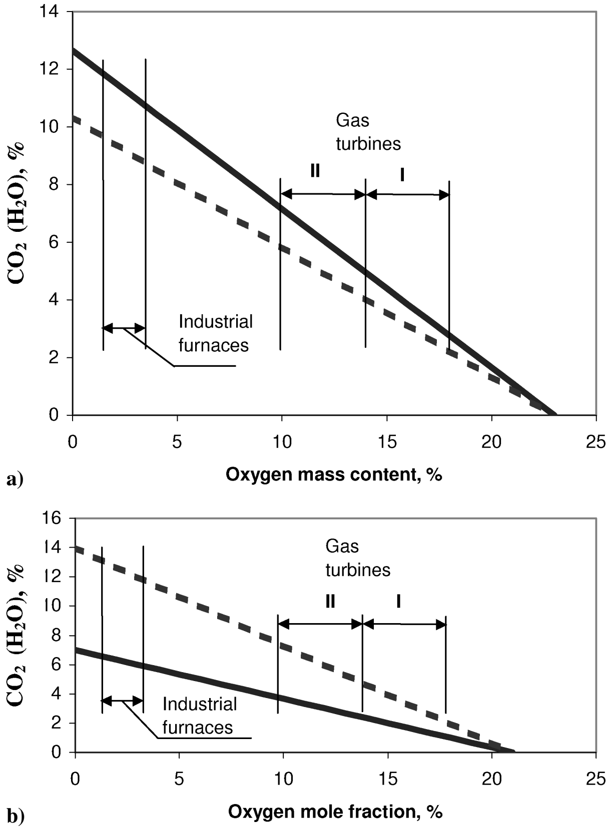

Fig. 1 O_2, CO_2, H_2O a) mass content and b) mole fraction at the recirculation zone for the combustor with recirculating gases; I, before combustion (stirring air); II, after combustion; ——, CO_2; and - - -, H_2O.

diluent is argon, helium, or nitrogen. The rates obtained by Peterson et al.[25,26] at high (above 1300 K) temperatures are

$$\tau = 1.26 \times 10^{-14} \exp(+32.7/RT)[CH_4]^{-0.02}[O_2]^{-1.2}\mathrm{s}$$

and at lower temperatures they have found:

$$\tau = 4.99 \times 10^{-14} \exp(+19.0/RT)[CH_4]^{-0.38}[O_2]^{-1.31}\mathrm{s}$$

As was pointed out by Lifshitz,[18] the oxygen has a promoting influence through the two branching reactions

$$CH_3 + O_2 \rightarrow CH_3O + O$$

$$O + CH_4 \rightarrow CH_3 + OH$$

The promoting effect (negative sign on the power dependency) means shortening of the ignition delay time (the explosion occurs faster). Contrarily, the fuel concentration has an inhibiting effect (positive sign on the power dependency) as a result of the competition on the H atoms between the fuel (methane) and oxygen in the main branching reaction of the combustion process:

$$H + CH_4 \rightarrow CH_3 + H_2$$

$$H + O_2 \rightarrow OH + O$$

The first reaction is only a propagation reaction. (One radical forms another radical.) The second reaction, besides being the most important combustion reaction in all fuel-oxygen systems, is also a branching reaction (One radical produces two radicals). Thus the increased concentration of fuel increases ignition delay time by reducing the concentration of H atoms. That explains the inhibiting effect encountered for methane in the Lifshitz equation[20,21] and in our experiments (see later). Petersen et al.[25,26] on the other hand found a promoting effect of the methane, and that is because they investigated very rich mixtures with methane concentrations between 30 and 50% ($\phi = 6$).

The diluent was not taken into account in the Lifshitz equation because it was found that for methane mixtures the effect of argon or nitrogen as a third body collider is negligible. Hence reactions where argon (or nitrogen) participates have very little effect on the ignition delay time although they are important for the procurement of other radical species.

To estimate the water and carbon-dioxide influence on the ignition delay, their relative content in the experiments was chosen close to the values in actual products of gas combustion (Fig. 1).

III. Experimental Section

In the present research two sets of mixtures were studied: 1) mixtures of methane-oxygen-nitrogen and 2) mixtures containing methane oxygen, nitrogen, CO_2, and H_2O with concentrations close to flameless oxidation combustion conditions (with recirculation of combustion products). The objective was to compare ignition delay time of these two sets.

Ultrapure Matheson methane 99.97% was used for the mixture together with 99% pure "oxygen stores" oxygen, nitrogen, and CO_2 95% pure. All of the gas mixtures were prepared manometrically. Distilled water was evaporated into a 70°C heated stainless-steel container. To prevent condensation of the water at the specific partial pressure used, the shock tube and all accessories were heated and maintained constantly at 70°C. Pure-grade helium was used as driving gas.

The experiments were performed behind reflected shock waves in a single-pulse shock tube. This instrument serves as a heater in the millisecond range, where the walls remain at room temperature (70°C in our case). The shock tube is made of a 4-m-long, 54-mm-diam stainless-steel tube. The driven section is 2.5 m long. Mylar diaphragms of different gauges were burst to generate the shock wave. Maximal time span behind the reflected shocks were 2 ms. The aerodynamic cooling achieved was $\sim 10^6$ K/s.

The sampling section, 0.25 long, contains three Kistler 603A piezoelectric transducers. Two transducers 0.20 m apart measure the shock velocity from which the temperature is calculated. A third piezoelectric gauge, located on the end plate, records the ignition delay time. The pressure outputs are fed to digital oscilloscopes that record 12 bit traces of 4098 points, each at 1-μs intervals.

The reflected shock temperatures were calculated using standard conservation equations and the ideal-gas equation of state assuming frozen chemistry. Thermodynamic data for all of the species were taken from Burcat's compilation.[28] The temperature uncertainty error is ± 20 K.

IV. Results and Discussion

Ignition delay in mixtures of methane-oxygen-nitrogen were compared with ignition delays in mixtures containing methane-oxygen-nitrogen, CO_2, and H_2O. In a preliminary study, argon and nitrogen were also used to find their influence on ignition. Table 1 lists all of the mixtures used in the experiments.

Figures 2–5 show the experimental points obtained for the different mixtures as log τ vs $1/T_5$ K. These mixtures were chosen to study the influence of argon and nitrogen for lean and stoichiometric mixtures on the ignition delay time (see Fig. 2). One can see that whenever O_2 and CH_4 contents are kept constant (almost) the replacement of Ar by N_2 does not affect ignition delay time. The results are in agreement with data obtained previously.[25,26] From Figs. 3, 4, and 5 it can be seen that there is no change of ignition delay if part of the nitrogen is replaced by CO_2 (Fig. 3), CO_2 and H_2O (Fig. 4), or all the diluents are compared to each other (Fig. 5).

Computer simulations were carried with the CHEMKIN SHOCK code.[23] The GRI 3.0 version mechanism and thermodynamics for methane, which contains 325 reactions, was used. It included C_1-C_3 species reacting with H and O atoms and molecules as well as reactions of oxygen and nitrogen. A second mechanism studied was Konnov's mechanism[29] (for model, see http://homepages.vub.ac.be/~akonnov/) with 1200 reactions and 127 species. One can see

Table 1 Composition of mixtures used for experimentation

Test No.	CH_4 mole, %	O_2 mole, %	N_2 mole, %	Ar mole, %	CO_2 mole, %	H_2O mole, %	ϕ	P_5 range, atm[a]	No of exper.
1	3.64	7.31	——	89.05	——	——	0.996	6.53–9.44	18
2	2.34	4.61	59	34	——	——	1.015	6.23–9.03	12
3	2.24	15.4	52.8	29.6	——	——	0.29	5.13–9.71	29
4	3.53	6.99	89.52	——	——	——	1.01	7.20–8.3	8
5	2.24	15.5	82.4	——	——	——	0.289	5.22–9.32	18
6	1.988	12.51	85.8	——	——	——	0.318	6.22–9.29	19
7	4.0	6.0	90.0	——	——	——	1.333	6.35–8.28	23
8	3.0	14.99	82.01	——	——	——	0.400	6.66–9.67	24
9	2.24	15.4	49.8	27.3	5.26	——	0.29	6.90–10.1	25
10	2.24	15.13	77.2	——	5.20	——	0.296	6.78–9.14	16
11	1.99	12.47	81.83	——	3.72	——	0.319	7.04–9.05	11
12	2.36	15.47	73.	——	5.15	4.02	0.305	6.55–8.99	17
13	2.24	15.47	73.2	——	5.15	4.02	0.29	6.80–8.9	22

[a] Notation $_5$ represents reflected shock conditions.

Table 2 Results of statistical calculation on experimental and modeled experiments

Data	σ	Statistics/ experiments	A	E_a, kcal/mol	α	β
Exp. data only	2	81/88	2.14×10^{-13}	36.5	0.278	−0.79
——	3	88/88	2.4×10^{-12}	36.7	0.211	−0.93
Exp. data +Hanson[a]	2	93/105	7.4×10^{-14}	35.7	0.112	−0.90
——	3	105/105	3.1×10^{-14}	35.5	0.209	−0.92
Modeling exp. data	2	35/37	5.62×10^{-13}	43.8	0.497	−1.31
——	3	36/37	4.17×10^{-14}	43.0	0.503	−1.35
Modeling exp. data +	2	39/50	6.6×10^{-16}	41.8	0.485	−1.4
Hanson's experiment[a]	3	48/50	7.8×10^{-16}	41.7	0.436	−1.45

[a] Ref. 26: Only data for high temperature (up to $T = 1300$) were taken from this paper.

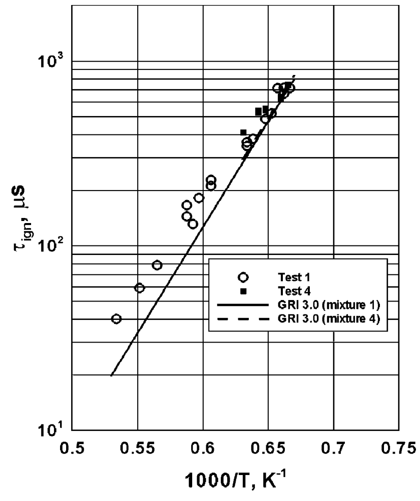

Fig. 2 Ignition delay time for stoichiometric mixtures. Comparison of methane oxidation in argon mixture 1 and nitrogen mixture 4. The modeling calculations of the two mixtures coincide.

(Fig. 3a) that the GRI mechanism is closer to the experimental data than Konnov's, but the slope of the simulation line calculated according to Konnov's mechanism fits better with our experimental data.

The partial replacement of N_2 by CO_2 (according to values determined by the flameless regime; see Fig. 1) does not affect the ignition delay time, as can be seen on experiments and simulation results (Fig. 3). On the other hand, experiments of Petersen et al.[25,26] at high pressures and low temperatures fit very well to our experiments even when treated statistically (see Table 2). A repeatable phenomenon seen in Figs. 2–5 is a deviation between experimental results and predictions at the high temperature range with significant better agreement at lower values. This is a well-known phenomenon when comparing experiments to published mechanisms. It means that the mechanism does not fit well the experimental evidence. However the relevant temperature range for practical combustion systems is the lower range, where good agreement was found.

In the present research we applied qualitatively the Lifshitz equation to our experiments, for comparison with earlier measurements. The mixtures, as shown in Table 1, were not prepared for calculations with the Lifshitz equation as there is not enough spread in the components concentrations; there is only a minute change of pressure, and there is absolutely no possibility to define the kinetic influence of the diluent.

The Lifshitz equation for the mixtures of methane-oxygen nitrogen as obtained from mixtures 5, 6, 7, and 8 is

$$\tau = 10^{-12.76\pm0.89}\exp(+36.4(\pm1.4)/RT)[\mathrm{CH_4}]^{0.31\pm0.14}[\mathrm{O_2}]^{-0.8\pm0.1}\,\mathrm{s}$$

This is very similar to the values found for methane and oxygen diluted in argon. The diluent is considered to have zero power dependence. The relatively low power dependence (∼ −0.8) of oxygen compared to the earlier findings of (∼ −1.) is also caused by the specific selection of the mixtures in this study and relatively low sensitivity of the oxygen change in the mixtures. A list of the all experiments of the different mixtures is given in the appendix.

A statistical treatment was performed on the calculated points, and the results are summed up in Table 2. The number of experiments that were used for statistical purposes from each group are listed. A few were rejected for being outside the assigned σ boundaries.

Figures 6 and 7 show the comparison of the Lifshitz equation with the kinetic model as is calculated by the Chemkin program. The figures show very clearly that the Lifshitz equation should not be extrapolated and used outside the experimental window. The agreement between the calculated and experimental data is mainly at 1300 K and pressure of 1.5 MPa as seen also on Figs. 2–5.

On Fig. 7 at the 1500 K region, it can be seen that the discrepancy between the experimental evidence and the predicted value of the mechanism is 100 μs. This is a value that engineers can accept

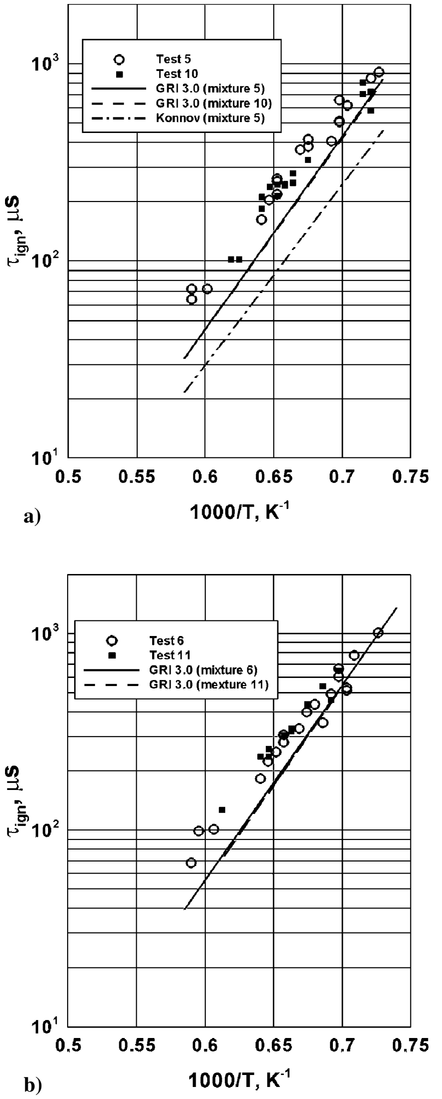

Fig. 3 Ignition delay time of mixtures containing nitrogen only (mixtures 5 and 6) and mixtures containing also CO_2 (10 and 11): a) the concentration of CH_4 and O_2 is kept constant in mixtures 5 and 10 at 2.24 and 15.5 mol%, respectively; and b) the concentration of CH_4 and O_2 is kept constant in mixtures 6 and 11 at 1.99 and 12.5 mol%, respectively.

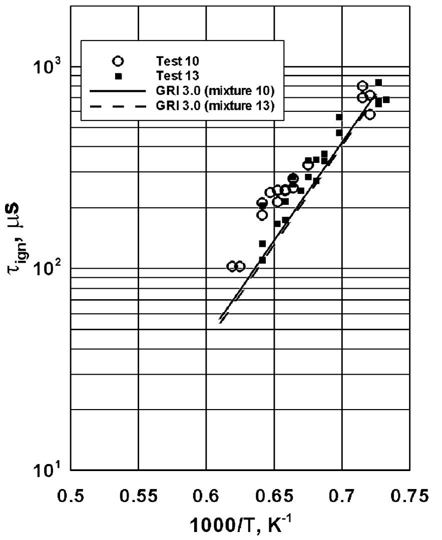

Fig. 4 Ignition delay time for mixture 10 containing CO_2 and 13 containing CO_2 and H_2O. The concentrations of CH_4 and O_2 are the same in both mixtures 2.24 and 15.5 mole%, respectively. No difference in behavior is found in in experiment and modeling as well.

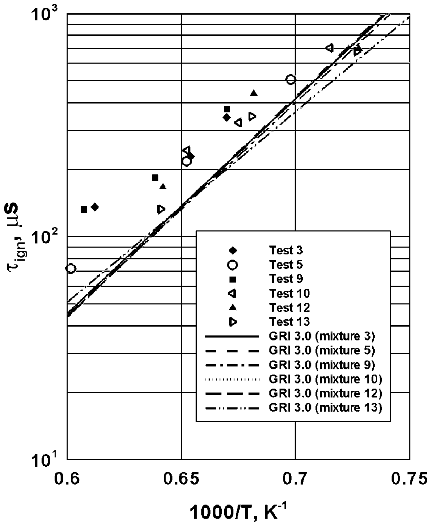

Fig. 5 Ignition delay time of all of the mixtures with the same concentration of methane and oxygen 2.24% CH_4 and 15.4% O_2 showing the same behavior regardless of the diluent, nitrogen, nitrogen and argon, nitrogen, argon and CO_2, or nitrogen CO_2 and H_2O. The calculations show the same behavior.

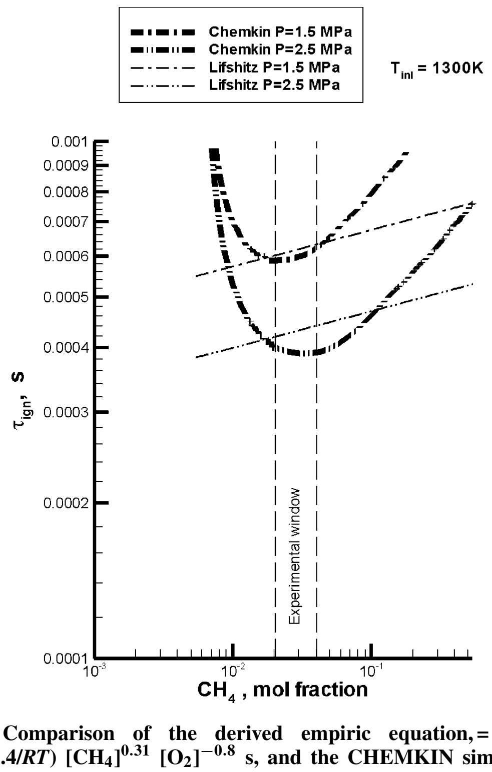

Fig. 6 Comparison of the derived empiric equation, $= 10^{-12.76}$ exp(+36.4/*RT*) $[CH_4]^{0.31}$ $[O_2]^{-0.8}$ s, and the CHEMKIN simulations results for ignition temperature of 1300 K. The oxygen concentration is constant at 21% (mole fraction).

because it is below the normal variations encountered during a real engine's work.

Lisianski et al.[22] have discussed the fact that the reaction $H + CO_2 \rightarrow CO + OH$ in a combustion process is competing on the H atoms with the branching main reaction of the combustion system $H + O_2 \rightarrow OH + O$. Therefore addition of CO_2 to the fuel as caused by reintroducing exhaust gases to the furnace should inhibit the combustion process. The GRI kinetic model does not confirm this behavior. The experiments show no ignition delay difference between the mixtures containing CO_2 and H_2O or without them, and the simulation calculations fully agree with this behavior as it is theoretically expected because CO_2 and H_2O will not react at low temperatures except.

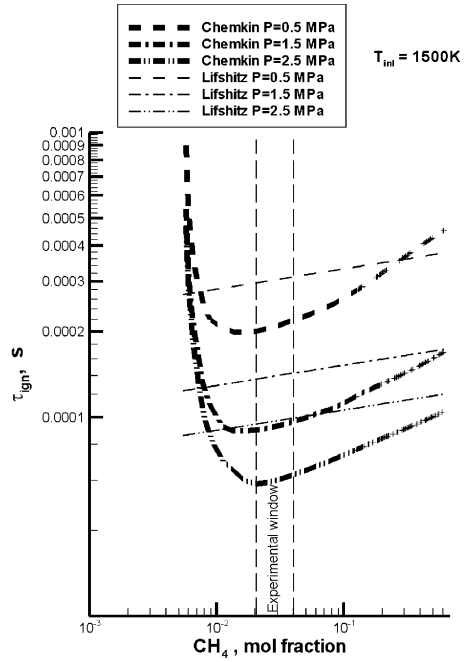

Fig. 7 Comparison of the derived empiric equation, = $10^{-12.76}$ exp(+36.4/*RT*) $[CH_4]^{0.31}$ $[O_2]^{-0.8}$ s, and CHEMKIN simulations for ignition temperature equal to 1500 K. The oxygen concentration is constant at 21% (mole fraction).

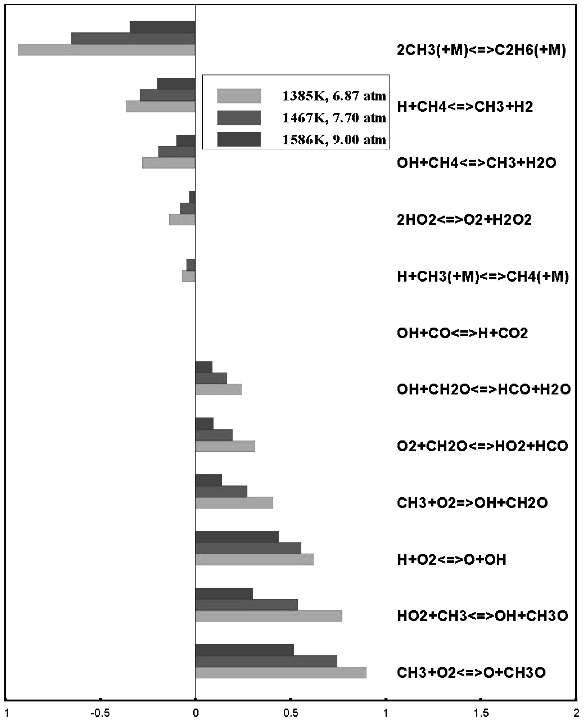

Fig. 8 Sensitivity analysis of the $CH_4 + O_2 + N_2 + CO_2 + H_2O$ model. The first 12 most sensitive reactions according to the GRI 3.0 mechanism. The sensitivities are calculated for the ignition temperature in a plug flow reactor.

Finally, in Fig. 8 the GRI 3.0 mechanism sensitivity of a plug flow reactor for the ignition stage is calculated. The 12 most sensitive reactions are presented according to the sensitivity analysis of CHEMKIN. Here as found also in the perfectly stirred reactor ignition zone, the CO + OH reaction is not important, and the two competing reactions are the methyl oxidation $CH_3 + O_2 \rightarrow O + CH_3O$ competing with the methyl recombination $2CH_3 + M \rightarrow C_2H_6 + M$. The same results were found in the flame area of the plug flow reactor.

V. Conclusions

Shock-tube and modeling investigation of ignition delay time was performed for mixtures simulating composition of the reacting gases, in an adiabatic combustor with internal recirculation. This parameter is relevant to the combustion stability of systems with reactants slightly diluted by combustion products.

The experiments were carried out in temperature range 1350–1800 K and pressure range 5–10 bars. Two mechanisms of reaction were used for modeling. The correlation between experimental data and the calculation results is in relative good agreement.

Different mixtures of CH_4-O_2-Ar-N_2 of various components concentrations were tested. It was found that replacing N_2 by Ar has not affected ignition delay time. Moreover, replacing the inert gases N_2 and Ar with CO_2 and H_2O was found to have negligible effect on ignition delay time. No other discrepancies between the recirculation produced in engines and the shock-tube experiments were found and neither in the calculated model results. Consequently it can be concluded that the combustion stability of the novel flameless oxidation gas-turbine combustor that utilizes internal recirculation of combustion products can benefit from the increase of the reactants temperature (by mixing with the hot products) without any deterioration caused by its chemical dilution.

Shock-tube experiments cannot duplicate the exact combustion conditions as the reactants in the tests at time zero do not include radicals. The radicals exist in real combustion environment, and their possible presence in the recirculated gases can shorten the ignition delay and improve stability of the combustion process.

Appendix: Operational Conditions of the Shock-Tube Experiments, Ignition in μs

Mixture 1: 3.64% CH_4 7.31% O_2 + Ar

No. of exp.	T_5, K	P_5, atm	τ_{ign}
1	1577	7.09	347
2	1701	8.01	144
3	1532	6.70	520
4	1510	6.53	715
5	1701	8.06	165
6	1650	7.69	210
7	1688	7.95	131
8	1770	8.55	78
9	1812	8.95	59
10	1873	9.44	40
11	1510	6.57	666
12	1500	6.49	713
13	1521	6.65	713
14	1650	7.65	227
15	1578	7.09	364
16	1675	7.80	181
17	1566	6.96	380
18	1543	6.82	484

Mixture 2: 2.24% CH4 15.4% O_2 52.76% N_2 + Ar

No. of exp.	T_5, K	P_5, atm	τ_{ign}
2an	1723	7.37	254
3an	1835	8.24	110
4an	1753	7.61	168
5an	1707	7.25	303
6an	1678	7.10	376
7an	1707	7.25	289
8an	1693	7.14	345
10an	1596	6.41	744
12an	1818	8.10	133
13an	1922	9.03	78
14an	1583	6.31	690
15an	1664	6.23	344

Mixture 3: 2.24% CH_4 15.4%O_2 52.8%N_2 + 29.6% Ar

No. of exp.	T_5, K	P_5, atm	τ_{ign}
35	1493	7.71	386
36	1459	7.43	383
37	1677	9.63	102
38	1554	8.32	193
39	1529	8.07	236
40	1459	7.43	350
41	1404	6.86	448
42	1394	6.76	640
43	1436	7.16	554
44	1554	8.32	161
45	1529	8.07	296
46	1567	8.51	192
47	1634	9.12	129
16	1677	7.35	75
17	1634	7.02	136
18	1505	6.02	280
19	1493	5.93	401
20	1493	5.93	343
22	1404	5.27	670
24	1677	7.35	102
25	1691	7.47	103
26	1482	7.72	437
27	1691	9.71	25
28	1394	6.76	772
29	1394	6.76	826
31	1529	8.13	230
32	1517	7.95	319
33	1517	8.01	316
34	1493	7.71	381

Mixture 4: 3.53% CH_4 6,99% $O_2 + N_2$

No. of exp.	T_5, K	P_5, atm	τ_{ign}
19n	1517	7.69	626
20n	1585	8.38	411
22n	1557	8.10	525
23n	1543	7.96	543
24n	1517	7.77	640
28n	1557	8.18	539
29n	1543	7.96	550
30n	1503	7.56	742

Mixture 5: 2.24% CH_4 15.7% $O_2 + N_2$

No. of exp.	T_5, K	P_5, atm	τ_{ign}
76	1533	7.71	253
77	1433	6.75	506
78	1546	7.84	204
79	1533	7.71	218
80	1560	7.98	162
81	1694	9.32	72
82	1533	7.71	261
83	1494	7.33	366
84	1433	6.82	655
85	1444	6.86	405
86	1481	7.21	413
88	1481	7.21	380
89	1387	6.39	846
90	1421	6.65	614
91	1433	6.82	512
92	1662	9.09	72
95	1376	6.23	906
98	1694	9.41	64

Mixture 6: 1.988% CH_4 12.51% $O_2 + N_2$

No. of exp.	T_5, K	P_5, atm	τ_{ign}
118	1422	6.62	513
120	1561	7.95	183
121	1521	7.56	281
123	1548	7.81	224
124	1648	8.81	101
125	1534	7.69	250
126	1679	9.13	99
127	1521	7.56	307
128	1470	7.07	437
129	1434	6.73	662
130	1445	6.85	493
131	1695	9.29	68
132	1434	6.73	605
133	1422	6.63	530
134	1458	7.03	353
135	1495	7.31	330
136	1483	7.26	401
137	1410	6.59	776
138	1377	6.22	1012

Mixture 7: 4.0% CH_4 6.0% $O_2 + N_2$

No. of exp.	T_5, K	P_5, atm	τ_{ign}
154	1534	7.93	584
155	1534	7.93	570
156	1590	6.80	415
157	1650	7.30	312
158	1697	7.79	204
159	1605	6.92	436
160	1764	8.28	104
161	1576	6.68	412
162	1730	7.98	126
163	1548	6.54	466
164	1650	7.39	280
165	1634	7.26	300
166	1521	6.40	711
167	1634	7.26	398
168	1548	6.54	467
169	1534	6.43	586
170	1576	6.77	505
171	1713	7.84	160
172	1576	6.68	425
173	1665	7.43	280
174	1713	7.94	151
175	1697	7.79	160
176	1534	6.35	635

Mixture 8: 3.0% CH_4 14.99% $O_2 + N_2$

No. of exp.	T_5, K	P_5, atm	τ_{ign}
177	1409	6.66	658
178	1520	7.73	245
179	1445	6.99	480
180	1507	7.60	298
183	1344	9.67	868
185	1409	8.66	600
186	1444	9.16	473
188	1387	8.38	733
189	1421	8.79	564
190	1445	9.09	486
191	1457	9.24	365
192	1481	9.55	278
193	1494	9.71	257
194	1469	9.40	353
195	1376	8.88	798
196	1421	9.47	445
197	1421	9.47	582
198	1433	9.63	466
199	1421	9.54	402
201	1421	9.40	410
202	1409	9.32	497
203	1421	9.40	436
204	1421	9.47	485
205	1387	8.96	751

Mixture 9: 2.24% CH_4 15.4% O_2 49.8% N_2 27.3% Ar 5.26% CO_2

No. of exp.	T_5, K	P_5, atm	τ_{ign}
48	1481	8.27	425
49	1492	8.40	374
50	1504	8.53	170
51	1528	8.79	174
52	1516	8.66	248
53	1492	8.40	388
54	1647	10.1	133
55	1633	9.95	107
56	1647	10.1	107
57	1414	7.57	588
58	1413	7.57	650
59	1413	7.57	565
60	1504	8.53	313
61	1424	7.68	539
62	1553	9.13	96
63	1579	7.26	174
64	1690	8.15	34
65	1541	6.87	240
66	1605	7.42	130
67	1605	7.49	158
68	1605	7.49	76
69	1566	7.15	184
70	1705	8.29	104
71	1661	7.90	81
72	1690	8.15	84

Mixture 10: 2.24%CH_4 15.13% O_2 77.2% N_2 5.20 CO_2

No. of exp.	T_5, K	P_5, atm	τ_{ign}
100	1519	8.11	242
101	1546	8.40	238
102	1616	9.14	102
103	1507	7.98	278
104	1398	6.89	702
106	1399	6.89	804
107	1533	8.33	213
108	1559	8.70	184
109	1533	8.33	244
110	1601	8.99	102
111	1507	7.98	250
112	1519	8.20	245
113	1481	7.72	325
114	1559	8.53	211
115	1387	6.78	721
117	1387	6.85	579

Mixture 11: 1.99% CH_4 12.47% O_2 81.83% N_2 3.72% CO_2

No. of exp.	T_5, K	P_5, atm	τ_{ign}
139	1434	7.04	648
141	1547	8.18	260
142	1633	9.05	127
143	1482	7.52	438
144	1446	7.16	460
145	1458	7.28	540
148	1547	8.26	238
150	1508	7.77	330
151	1508	7.77	322
152	1521	7.90	306
153	1561	8.31	238

Mixture 12: 2.36% CH_4 15.47% O_2 73% N_2 5.15% CO_2 4.02% H_2O

No. of exp.	T_5, K	P_5, atm	τ_{ign}
31	1396	7.06	654
32	1467	7.71	439
33	1531	8.39	212
34	1558	8.68	167
35	1572	8.83	167
36	1385	6.88	696
37	1455	7.59	476
38	1385	6.88	630
39	1408	7.10	738
42	1353	6.55	740
44	1443	7.46	336
45	1586	8.99	113
46	1518	8.25	260
47	1558	8.68	162
48	1374	6.77	700
49	1455	7.59	437
50	1467	7.71	374

Mixture 13: 2.24% CH_4 15.47% O_2 73.2% N_2 5.15% CO_2 4.02% H_2O

No. of exp.	T_5, K	P_5, atm	τ_{ign}
51	1433	7.50	470
54	1376	8.88	656
55	1365	8.67	688
56	1376	8.81	677
57	1507	8.21	284
58	1481	7.86	285
59	1560	8.70	110
60	1560	8.70	133
61	1520	8.35	215
62	1469	7.81	273
63	1494	8.07	243
64	1457	7.68	341
65	1433	7.35	565
66	1457	7.60	370
67	1520	8.26	174
68	1469	7.80	347
69	1560	8.70	205
70	1376	6.85	837
71	1507	8.21	262
72	1533	8.49	167
73	1469	7.73	404
74	1481	7.94	344

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