1. Detailed Experimental Methods

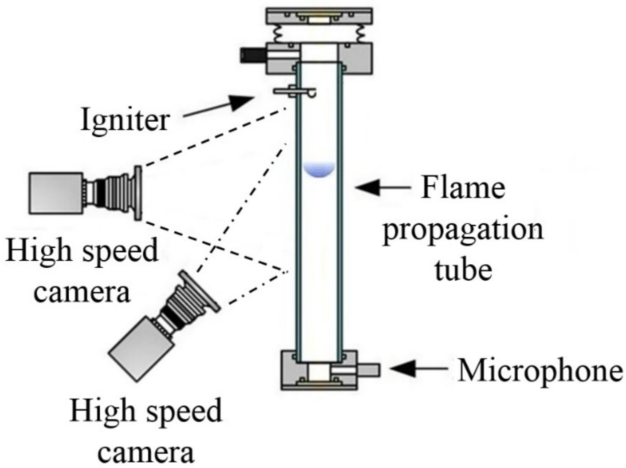

Fig. 1 Schematic representation of the experimental apparatus.

The experimental apparatus is schematically outlined in Fig. 1. The propagation tube (transparent acrylic tube, 50 mm inner diameter, 711 mm length) was fixed vertically and charged with the tested gas at atmospheric pressure. A point ignition system using a spark plug was located near the upper end of the tube. When the spark igniter was activated, the top lid was simultaneously opened by an automatic opening system of the top lid of the tube powered by an electromagnet and a mechanical spring. The time-dependent behaviors of the downward-propagating flame were recorded by two high-speed cameras. One of the high-speed cameras (FASTCAM Mini UX100 recording at 2000 fps) captured images from side view of the tube. Another high-speed camera (nac HSV-500C3 recording at 500 fps) captured bottom view of the propagating flame in inclined angle as shown in Fig.1. Temporal pressure variation was measured with a PCB Piezotronics 106B52 dynamic pressure sensor located at the bottom end of the tube at a sampling rate of 10 kHz.

In this study, the premix gas composed of propane, oxygen and nitrogen were set at atmospheric pressure in the tube as shown in Table 1. We selected equivalence ratios of 0.8 and 1.2 to change *Le*. To vary laminar burning velocities with the fixed equivalence ratio, we changed dilution ratio, $D = \frac{O_2}{O_2 + N_2 \text{ or } CO_2}$. The flame properties (e.g., laminar burning velocities, S_L, speed of sound, c, and adiabatic flame temperatures, T_b) were calculated using CHEMKIN (Premix code/ USC II) [1]. The Zel'dovich number could be calculated by $\beta = E(T_b - T_u)/(RT_b^2)$. Here, E is the activation energy, R is the gas constant, and the subscripts u and b identify unburned and burned mixtures, respectively. Peters and Williams [2]

Table 1 Detailed gas properties for the tested gas.

Mix.	C_3H_8	O_2	N_2	Φ	S_L (cm/s)	T_b (K)	βM	Le	$\beta(Le$-1)	Regime
1	0.027	0.169	0.804	0.8	17.5	1833	0.0058	1.86	9.82	I
2	0.028	0.175	0.797		20.0	1881	0.0065		9.72	I
3	0.029	0.182	0.789		22.5	1927	0.0074		9.76	I
4	0.030	0.188	0.782		25.0	1969	0.0083		9.84	II
5	0.031	0.194	0.775		27.5	2009	0.0092		9.85	IV
6	0.032	0.200	0.768		30.0	2046	0.0101		9.78	IV
7	0.033	0.206	0.762		32.5	2082	0.0110		9.84	IV
8	0.034	0.211	0.755		35.0	2114	0.0120		9.89	III-2
9	0.035	0.217	0.749		37.5	2145	0.0129		9.92	III-2
10	0.035	0.222	0.743		40.0	2175	0.0139		10.03	V
11	0.036	0.227	0.737		42.5	2203	0.0151		10.17	V
12	0.033	0.135	0.832	1.2	7.5	1705	0.0038	1.04	0.73	II
13	0.034	0.142	0.823		10.0	1761	0.0046		0.63	II
14	0.036	0.149	0.816		12.5	1812	0.0053		0.56	III-1
15	0.037	0.155	0.808		15.0	1859	0.0060		0.52	III-1
16	0.039	0.161	0.801		17.5	1905	0.0066		0.48	IV
17	0.040	0.166	0.794		20.0	1948	0.0073		0.45	IV
18	0.041	0.172	0.787		22.5	1990	0.0079		0.41	IV
19	0.042	0.177	0.781		25.0	2030	0.0086		0.40	V
20	0.046	0.192	0.761		32.5	2139	0.0108		0.36	V
21	0.051	0.212	0.737		42.5	2268	0.0138		0.32	V

defined the activation energy as $\frac{E}{R} = -\frac{\mathrm{d}2[\ln(\rho_u S_L)]}{\mathrm{d}(1/T_b)}$ where ρ is density of gas mixture. The Mach number, M, which appears in table1, is defined as $M = S_L/c$. Lewis number was given by the ratio of the thermal diffusivity of mixture to the mass diffusivity of deficient gas species, $Le = \alpha/D_{dif}$. We selected two different *Le* for $Le \approx 1.85$ from Mix. 1–11 and $Le \approx 1.04$ from Mix. 12–21. To observe pulsating instability, we conducted experiments with various reduced *Le*, $\beta(Le\text{-}1)$, which can be varied by different equivalence ratios and laminar burning velocities.

Reference

[1] Z. Qin, V. Lissianski, H. Yang, W. C. Gardiner, S. G., Davis, H. Wang, Proc. Combust. Inst. 28 (2000) 1663-1669.

[2] N. Peters, F. A. Williams, Combust. Flame 68 (1987) 185-207.