Brief Communication

Observation of liquid phase material in methane–air laminar diffusion flame soot experiments above 60 atmospheres

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1. Introduction

An interesting occurrence was observed during the study of soot formation in laminar diffusion flames of methane and air at pressures exceeding 60 atm. The laminar flame established on a circular nozzle-type burner at 60 atm and higher pressures was extinguished by itself and immediately after a liquid substance emerged from the fuel nozzle. To our knowledge, no observations were reported on liquid formation during methane combustion at elevated pressures. The aim of this brief communication is to report the observation of this anomalous occurrence.

The apparatus used for the present investigation is a high pressure combustion chamber with an inner diameter of 0.24 m and a height of 0.60 m and described in detail previously [1]. The co-flow burner has a fuel nozzle diameter of 3.06 mm and an air nozzle diameter of 25 mm. A constant methane mass flow rate of 0.55 mg/s that corresponds to a carbon mass flow rate of 0.412 mg/s was used for all the pressures. Soot and temperature measurement method used in this study is described in detail in [1,2].

2. Anomalous observation and discussion

A stable laminar diffusion flame is established at atmospheric pressure first before pressurizing the chamber. At the chamber pressure of 60 atm, an interesting occurrence was noticed. While nitrogen was employed as the viewport purge gas to remove water condensation on the quartz windows, the flame was extinguished and immediately a clear and transparent liquid bubble emerged out of the fuel nozzle, Fig. 1.

Similarly at 80 atm the fuel flow rate was increased temporarily and small liquid bubbles emerged and spilled onto the side of the fuel nozzle. Typical images depicting the time sequence of the process in chronological order are shown in Fig. 2 for the chamber

pressure of 80 atm. The images in Fig. 2b and c show a rapid increase in the visible flame height just before it is rapidly decreased to extinction as seen in Fig. 2e. Immediately after, a liquid bubble emerged out of the nozzle and continued to grow in size as shown in Fig. 2f–h. As the bubble continued to expand, free-floating particulates were observed to float on the surface of the bubble. With the fuel continuing to flow, the liquid bubble bursted and a new bubble emerged from the fuel nozzle. This cycle was repeated often several times or until the fuel flow was stopped. When the pressure was decreased to atmospheric the liquid was completely evaporated and solid deposits on the surfaces of the fuel nozzle were observed.

Images in Fig. 3 show the liquid in concert with the flame and affecting the overall geometrical structure of the flame. In Fig. 3a, the combustion chamber was pressurized with air and the flame was established for 30 min prior to the onset of the liquid formation where it begins to disturb the flame stability. The liquid acts as a base to support a smaller secondary flame from the same nozzle, Fig. 3b. Once the liquid is observed just at the tip of the fuel
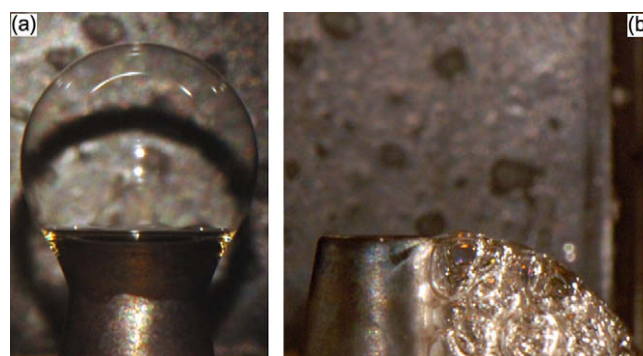

Fig. 1. Liquid formation at pressures above 60 atm. (a) $P = 60$ atm. Clear and transparent liquid bubble with diameter 6.5 mm. (b) $P = 90$ atm. Small broken liquid bubbles spilled to the side of the fuel nozzle.

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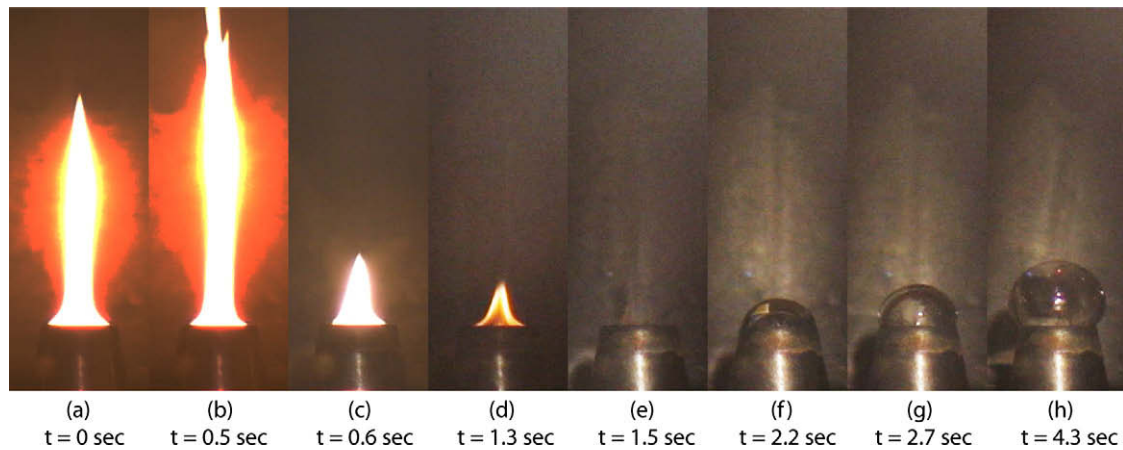 E-mail address: ogulder@utias.utoronto.ca (Ö.L. Gülder).

Fig. 2. Sequence of images showing the development of liquid bubble at P = 80 atm. (a) The usual appearance of the diffusion flame at 80 atm with the visible flame height of about 9 mm, and the flame is thermally stabilized; (b and c) rapid increase then decrease in flame height; (d and e) flame extinction; (f–h) formation of liquid bubble and its growth.
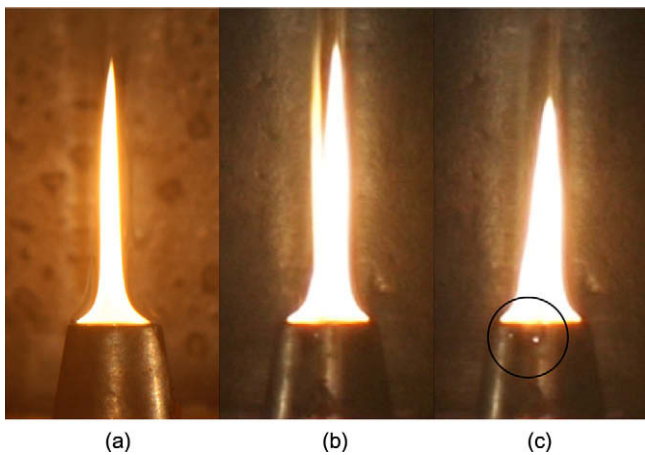

Fig. 3. Flame and liquid phase material at 90 atm. (a) Flame at P = 90 atm with height 8.8 mm and thermally stabilized. (b) Onset of liquid formation and burn-off (t = 0 s). (c) Liquid bubble on the side of the nozzle (indicated by a circle) and change in flame geometry (t < 10 s).

A possible explanation can be provided by the fact that the water vapor in the combustion products could be creeping inside the fuel nozzle and mixing with the supercritical methane [3]. At 60 atm and above, the methane is supercritical and can form liquid hydrate at the right temperature in the presence of water. For example, equilibrium water mole fraction in the supercritical methane fluid is about 35% at 60 atm and 478 K [4]. These temperatures are possible especially at high pressures when the soot formation region is very close to the burner rim and soot concentrations are much higher than atmospheric. Thermodynamic data based on calculations and presented in [4] show that the liquid observed is liquid methane hydrate. Once a sufficient amount of liquid methane hydrate is formed the liquid emerges from the fuel tube and the flame retracts rapidly towards the fuel nozzle to extinction, Fig. 2.

References

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nozzle, the visible flame height decreases and the flame widens near its base, Fig. 3c. The liquid is then consumed and the flame regains its original shape only until a new liquid bubble is formed to repeat the process.

It should be noted that liquid phase occurrence was not a rare event but it consistently occurred at 60 atm and above. It was repeatable except the onset of it after the set pressure was reached.