Self-ignition and flame propagation of pressurized hydrogen released through tubes

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Abstract

The spontaneous ignition of hydrogen released from the high pressure tank into the downstream pipes with different lengths varied from 0.3m to 2.2m has been investigated experimentally. In this study, the development of shock wave was recorded by pressure sensors and photoelectric sensors were used to confirm the presence of a flame in the pipe. In addition，the development of jet flame was recorded by high-speed camera and IR camera. The results show that the minimal release pressure in different tube when self-ignition of hydrogen occurred could decrease first and then increase with the increase of the aspect of pipe. And the minimum release pressure of hydrogen self-ignition was 3.87MPa. When the flame of self-ignition hydrogen spouted out of the tube, Mach disk was observed. The method of CFD was adopted. The development of shock wave at the tube exit was reproduced and structures as barrel shock, the reflected shock and the Mach disk are presented. Because of these special structures, the flame at the nozzle is briefly extinguished and re-ignited. At the same time, the complete development process of the jet flame was recorded, including the formation and separation of the spherical flame. The flame structure exhibits three typical levels before the hemispherical flame separation.

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Introduction

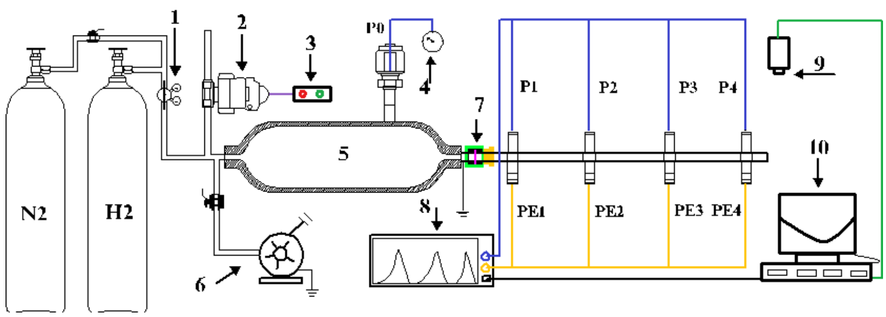
Hydrogen has high calorific value of combustion, and is one of the most important clean energy sources. And many developed countries believe that it is necessary to develop hydrogen fuel cells and hydrogen industry (1). However, the minimum ignition energy of hydrogen is 0.017 mJ (2). The limit of hydrogen combustion is very wide, ranging from 4 to 75 %. And Hydrogen can easily cause fire and explosion when it leaks. When high-pressure hydrogen gas is transported through pipelines or stored in storage tanks, hydrogen leakage may occur in the piping connection and other locations. More and more countries attach importance to the development of hydrogen energy in recent years. Therefore, it is significant to accumulate hydrogen safety data through some release experiments to prevent hydrogen accidents.

Numerous experiments found that a sudden release of high-pressure hydrogen could form the shock wave at the high-speed jet front, and the high pressure and temperature generated by the shock wave would heat the hydrogen-air mixture layer so that hydrogen ignition may occur at the mixture layer. In 1973, Wójcicki (3) firstly found that hydrogen spontaneously ignited in shock tube experiments, although the ambient temperature was lower than the hydrogen self-ignition temperature. Mogi (4, 5) applied the pipes of different lengths between 3mm and 300mm, and they found that the initial release pressure of hydrogen self-ignition could decrease with the increase of the pipeline length. And they thought that the increased pipeline length created the great condition for hydrogen self-ignition, such as full mixing of hydrogen and air and maintenance of high temperature. However, this paper found the initial release pressure could rise in the longer length of pipes in this experiment, and this variety may be related to half-constrained space in the pipeline. Duan (6, 7, 8) found that the higher release pressure of hydrogen, the longer length and the smaller internal diameter of pipes could promote hydrogen spontaneous combustion, and the closer of hydrogen self-ignition located to rupture discs. But, the longest length of pipes was

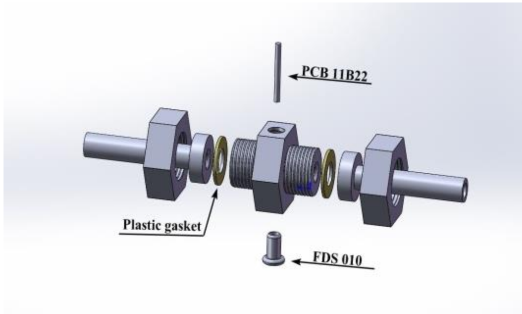
360mm in their experiments. With the increase of the pipe length, the pressure changes became more complicated inside the pipe, especially the influence of the disc multi-step bursting and some shock reflections. Kitabayashi (9) studied the relationship between the different lengths of pipes and the critical pressure of hydrogen self-ignition through the different lengths of 0.1-4.2m. Nonetheless, they did not employ the photoelectric sensors in experiments. And the photoelectric sensors can monitor the location of hydrogen self-ignition inside the pipe.

This experiment use different length pipes to research high-pressure hydrogen self-ignition. And the pressure sensor was used to measure the pressure of the pipe wall, and the photoelectric sensor was employed to detect phenomenon and location of hydrogen self-ignition in pipes after the release of hydrogen from the gas tank. Through a large number of experiments, we studied the process of hydrogen spontaneous combustion inside the pipeline and the formation of a jet flame outside the tube. Collect additional data and complement previous research. Firstly, this paper studied the self-ignition of hydrogen in different tube. And author analyzed the relationship between the minimum pipe wall pressure of hydrogen self-ignition and the aspect ratio of pipe. And through the combination of experiment and simulation, the transformation process of flame at the nozzle is studied. The development of transient flames was studied jointly by high-speed camera and IR camera.

Experimental setup



(a)



(b)

Fig. 1 Experimental apparatus：(a) Schematic of the experimental setup (1) Regulator valve, (2) Pneumatic valve, (3) Controller, (4) Pressure gauge, (5) High-pressure tank, (6) Vacuum system, (7) Rupture disc, (8) Data recorder, (9) high-speed video camera, (10) Computer, (b) The connection of different short tubes

The experimental apparatus is shown in Figure 1. In the experiment, the hydrogen in the gas cylinder upstream of the pipeline was continuously filled into the high pressure tank, causing the tank pressure gradually rise until the rupture disc broke, and finally the high pressure hydrogen was suddenly released into the downstream pipeline. As shown in Fig.1（a）, the experimental device contains six parts: gas supply system, high pressure tank, the downstream pipeline, the holder with a rupture disc, data acquisition system and control system. A pneumatic valve is installed between the cylinder and the storage tank. The downstream pipe has a diameter of 10 mm. Each pipe is made up of three short length pipes of the same length. Fig. 1(b) shows the schematic of the connection of different short tubes. Pressure and photoelectric sensors are installed between each short tube. The holder with a rupture disc is installed between the downstream pipe and the high pressure tank, whose position is 65 mm from the first sensor. The rupture disc made of nickel has a “cross symbol” weakening groove, whose burst pressure varies 2MPa to 9MPa. The development of shock wave was recorded by pressure sensors and photoelectric sensors were used to confirm the presence of a flame in the pipe. A high-speed camera is placed outside the tube to record the flame development process outside the tube.

Result and discussion

Self-ignition and shock waves in the tube

Table 1 different initial conditions of high pressure hydrogen release experiments

No.	Pipe length (mm)	Burst pressure (MPa)	Mean velocity of the shock wave (m/s)	The highest pipe wall pressure (MPa)	Positions of light signal detection
1	1200	3.98	1098	2.58	No
2	1200	5.01	1152	2.40	L2+L3+L4
3	1200	6.04	1215	3.60	L2+L3+L4
4	1700	2.89	1003	1.84	No
5	1700	3.70	1105	2.89	L3+L4
6	1700	5.02	1181	3.72	L2+L3+L4
7	1700	5.98	1207	3.80	L1+L2+L3+L4

The conditions in the experiment are listed in Table 1, including pipe length, burst pressure, the mean velocity of the shock wave, the highest pipe wall pressure and the position of light signal detection. The average velocity of shock wave is calculated by the distance between sensors and the time difference between the shock waves captured by pressure sensors. The highest pipe wall pressure is the maximum value of the pressure curve monitored by the pressure sensor. It can be inferred from Table 1 that the burst pressure has great influence on spontaneous ignition of hydrogen and the position of light signal detection. Under the same length of pipeline, as the burst pressure increases, the average shock velocity grows and the possibility of spontaneous combustion of high-pressure hydrogen increases. At the high burst pressures, strong shock wave is generated inside the pipe, which can be inferred from the average shock velocity and the maximum wall pressure in Table 1. The stronger the shock wave, the higher the temperature in the area behind it. Therefore, the strong shock wave heats the mixed area of hydrogen and air. Once the local temperature reaches the auto-ignition point of hydrogen, high pressure hydrogen will spontaneously ignite inside the pipe. Furthermore, in different length of tube, the lowest burst pressure is different when spontaneous combustion occurs inside the pipeline. In the 1200 mm pipes, the photoelectric sensor did not capture the signal under the burst pressure of 4MPa, indicating that there was no spontaneous combustion, but auto-ignition occurred in the 1700 mm pipes.

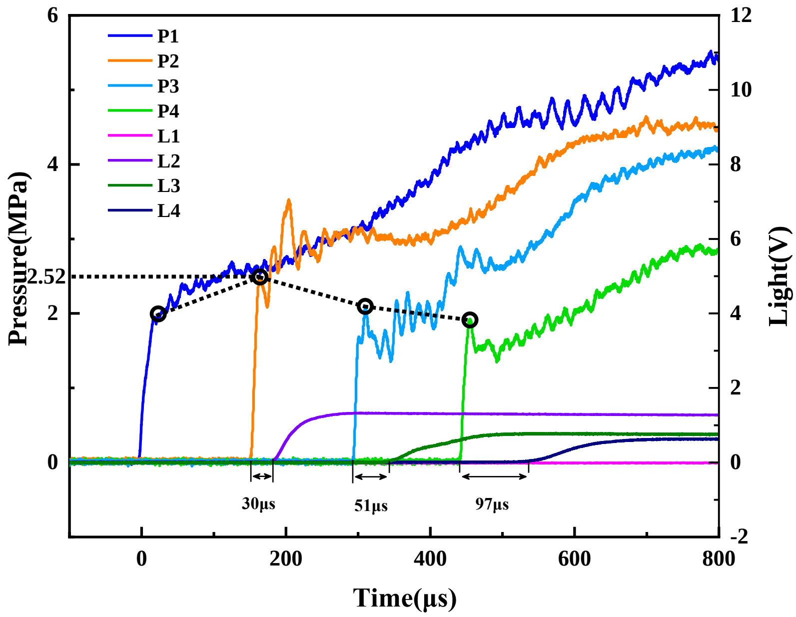
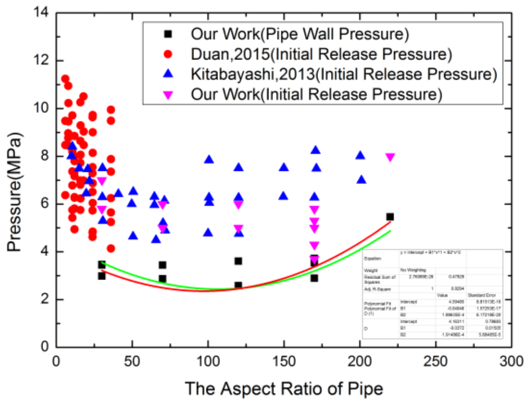
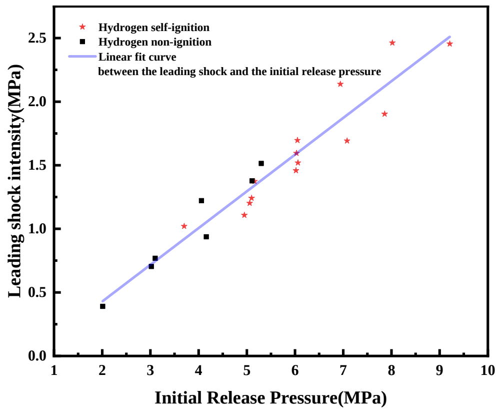


Fig. 2 Typical curves of pressure and photoelectric signal

In Figure 2, the maximum leading shock was detected at P2. Because the intensity of leading shock would fluctuate. The mutual interference of the shockwave in the propagation process increased the intensity of leading shock, and energy dissipation because of fluid viscosity and wall friction reduced the intensity of the leading shock (compared P1 with P4 under one condition). Therefore, the leading shock was relatively stable under the combined action of the reinforcing effect and the weakening effect. In Figure 2, L2 (photoelectric sensor) first received the photoelectric signal, which shows that the ignition position was between L1 and L2 inside the tube. By comparing the photoelectric signal with the pressure signal, it was found that the leading shock should be in front of the ignition position. Moreover, the interval between the leading shock signal and the photoelectric signal was lengthened gradually, indicating that the mixed zone enlarged gradually and the shock wave speed was faster than the flame inside the tube.





(a) (b)

Fig.3 （a）The self-ignition map showing the relationship between the aspect of pipe and the burst pressure；（b） Relationship between initial release pressure and intensity of leading shock

Fig. 3（a）shows a map of self-ignition created from the experimental results using the extension tube of various lengths and different diaphragm burst pressures. A fitting curve is roughly given to show the minimum release pressure that causes spontaneous ignition in different tubes. From the figure, we can conclude that as the aspect ratio of pipe increases, the release pressure of hydrogen spontaneous combustion undergoes a process of decreasing first and then increasing. The most important result of this study is the existence of a minimum in the dependence of the diaphragm burst pressure. The minimum release pressure to induce the spontaneous ignition is 3.87MPa with the tube length of 1.7m. Fig.3（b）show the relationship between the initial release pressure and the leading shock intensity. The red point indicates the intensity of the leading shock wave when spontaneous combustion occurs, and the black point indicates the intensity of the leading shock wave when spontaneous combustion does not occur. The leading shock intensity was derived from the stable leading shock intensity that was detected by the pressure sensor. In Fig.3（b）, it can be found that the greater the initial release pressure, the greater the leading shock intensity, and the leading shock intensity was distributed up and down the fitting curve. It could also be seen from the Fig.3（b） that high-pressure hydrogen was not prone to self-ignition when the leading shock intensity was low; and when the leading shock intensity was strong, the high-pressure hydrogen was more likely to spontaneously ignite inside the tube. And the minimum leading shock intensity caused hydrogen self-ignition in this experiment was approximately 1.00 MPa. As the initial release pressure increased, the leading shock intensity increased, and the flow in the hydrogen-air mixed zone and the hydrogen jet zone was enhanced, and hydrogen and air mixed more intensely at the contact surface, and shock compression and friction heat generation would be enhanced, and eventually it would promote the hydrogen self-ignition.

The development of flame out of the tube

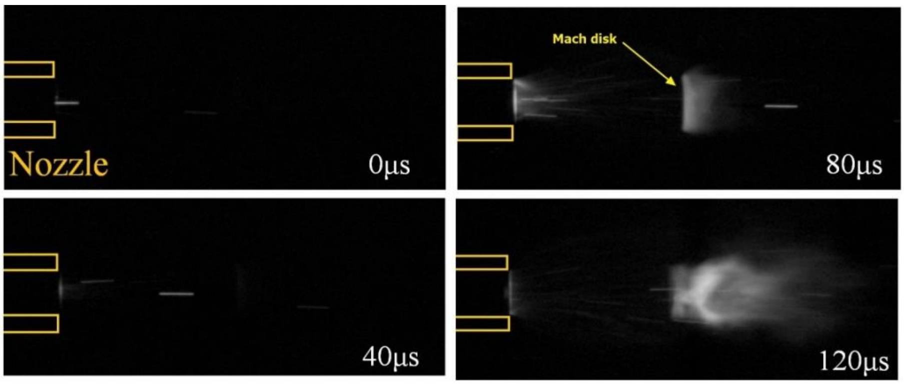


Fig. 4 Transition of spontaneous ignition into a jet fire

To study the mechanism how the flame transforms into a jet fire a jet fire at the exit of the tube, a high-speed camera is used to obtain the visual data. Fig.4 shows some the direct images（130000fps）of flame at a pipe with a diameter of 10mm and a length of 2200mm. It can be seen that the flame that originally emerged from the outlet of the pipe is close to the center of the tube, and the flame has a certain length. And then, the flame propagated downstream. As time goes on, the bright area of the nozzle increases gradually and its width is equal to the diameter of the pipe, indicating that the flame is gradually stable at the nozzle. At the same time, the Mach disk appeared at a certain distance from the nozzle, and a flat flame was generated at the rear of the Mach disk. Between the Mach disk and the nozzle flame, the combustion is weak and there is substantially no flame. When the hydrogen jet passes through the Mach disk, it is re-ignited. It can be seen from picture that the burning after the Mach disk is more intense. As time goes on, the flame at the nozzle disappeared and the flame ignited by the Mach disk propagates downward, eventually becoming a jet fire.

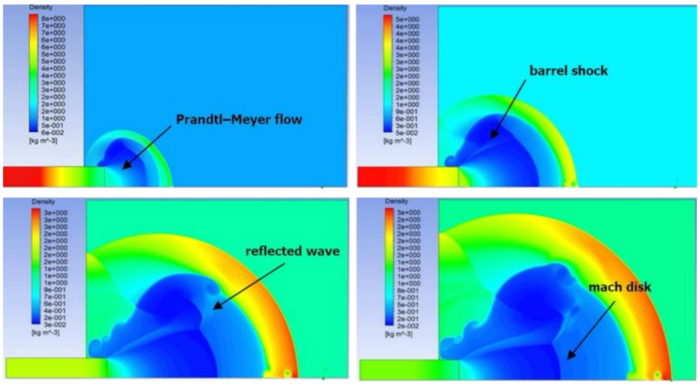


Fig. 5 Development of shock wave at tube exit

To further analyze the flame development at nozzle and understand the structure of the hydrogen flow field, we simulated the development of shock waves at the nozzle. The LES model was used in this study. The energy equation and the component transport equation were solved simultaneously. A two-dimensional symmetric structure was used. The simulation reproduces the development of the shock wave at the nozzle. Fig.5 shows the development of shock wave at tube exit. When the high-pressure hydrogen jet suddenly enters the enlarged space, due to the influence of the Prandtl–Meyer flow, a triangular expansion fan is formed at the nozzle, and the flow field parameters on both sides of the expansion fan change greatly, which results in a triangular expansion flame at tube exit as shown in Fig.4 when t=80 μ s. As the high pressure hydrogen jet expands further, the barrel shock, reflected shock, and the Mach disks occur in sequence. The flow field parameters on both sides of the Mach disk change drastically. After the airflow passes through the Mach disk, the pressure increases, the speed decays, and the temperature rises, which causes the jet to be re-ignited. As can be seen from Figure 4, after the flame comes out of the tube, it does not extinguish immediately, but decays through a triangular boundary and reignites after the Mach disk. It can be seen from the simulation that there are two faces between the expansion fan and the Mach disk which parameters changes drastically, and the flame changes in the middle.

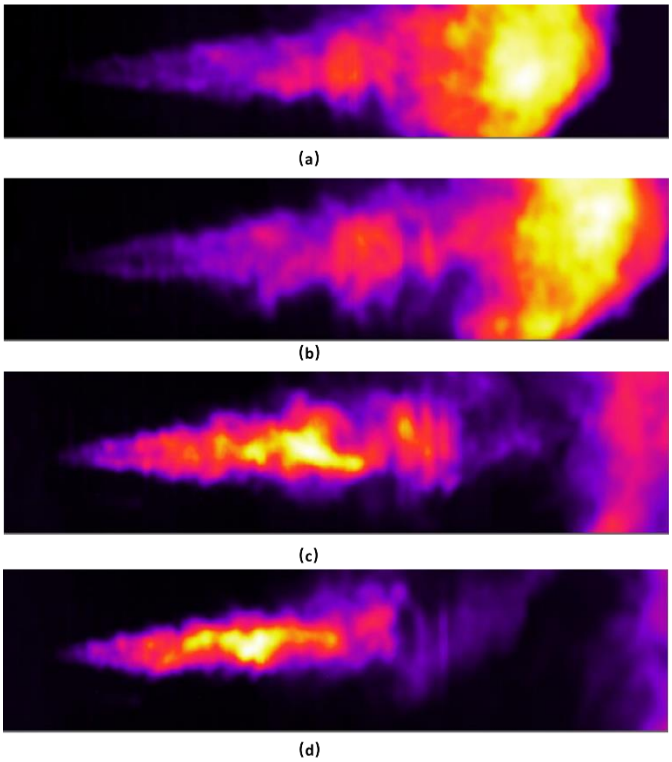


Fig. 6 Propagation of hydrogen jet flame（IR images）

When the spontaneous flame of hydrogen was successfully converted to the jet fire, its entire development process was recorded. Fig.6 shows a series of images obtained during the flame propagation process. When high pressure hydrogen is ejected from the nozzle, it rapidly expands along the centerline. The flame boundary presents a certain angle with the centerline, and the overall structure of the flame is tapered as shown in Fig. 6（b）. Due to the action of the boundary layer, the center of the jet is faster and the boundary speed is lower, which can be seen intuitively from the animation captured by camera. This results in a speed difference on the flame front, leading to a hemispherical structure at the front of the flame, as the flame propagates downstream to a certain distance. The bright area appears at the front of the flame, and the diameter of the spherical flame increases first as shown in Fig. 6（b）. Then the connection between the flame head and the flame body is weakened. In the experiment, a huge explosion sound was heard, and it was speculated that an explosion of a mixture of hydrogen and oxygen occurred at the front end of the flame. Subsequently, due to the closure of the upstream valve and the constant attenuation of pressure in the tank, the spherical flame region is separated from the flame vertebral body. From the Fig. 6（d），the flame separated from the jet flame continuously propagate forward，and eventually disappeared. Judging from the IR images, the front end flame burns fiercely, the temperature is higher, and the propagation distance is farther. It is more destructive to surrounding objects.

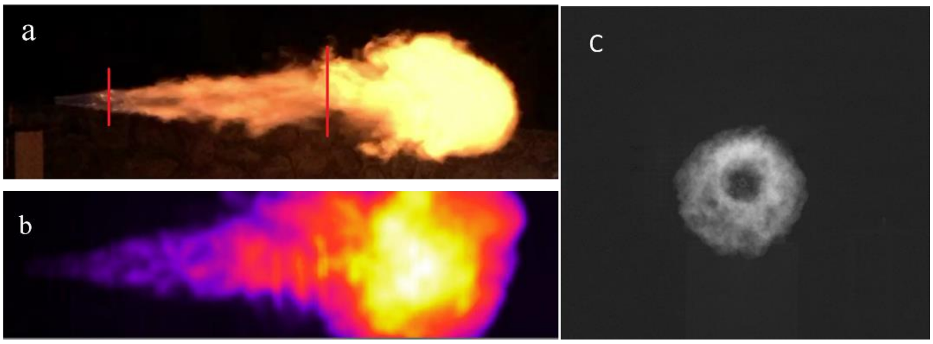


Fig. 7 Structure of jet flame

Figure 7（a） was taken by the camera, Figure7（b） was taken by the infrared which were placed at the side of experimental setup, .Figure 7（c） was taken by the high-speed camera set in front of the flame. It can be seen from Figure 7（a） that the flame can be divided into three distinct segments. The root of flame burned weakly and was similar to laminar flow. In the middle of the flame, a severe turbulent combustion zone was observed, the boundary layer flame was considerably wrinkled, and the flame gradually thickened. At the head of the flame, the flame burns fiercely and the connection to the flame body is weakened. From the middle of the flame in Figure 7（a） we can see that at the core of the jet, the flame burns weakly. We can see from the picture taken by the flame head camera A non-stick core (hydrogen was not mixed with air) was observed at the centerline of the jet.

Conclusion

In this study，we investigated the characteristics of self-ignition of hydrogen releasing from high-pressure tank by changing the pipe length and burst pressure. The photoelectric sensor signals confirm the spontaneous combustion of hydrogen in the pipeline.

(1) The shock wave propagating in a small-diameter pipeline undergoes a process of increasing first and then decreasing. The propagation process is a non-isentropic process. And when spontaneous combustion occurs, the speed of shock wave is higher than that of the flame.

(2) The initial release pressure of hydrogen spontaneous combustion undergoes a process of increasing first and then decreasing as the length-to-diameter ratio of the pipe increases.

(3) In the process of flame transition to jet fire, the Mach disk plays an important role. The hydrogen jet is re-ignited after the Mach disk to form a flat flame. The development of shock wave at the tube exit was simulated and structures as barrel shock, the reflected shock and the Mach disk are presented. The flame at the nozzle is triangular by the influence of the Prandtl-Meyer flow.

(4) The jet fire presents a typical three-stage structure with an unburned area in the center of the flame.

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