# FUNDERMENTAL STUDY ON ACCIDENTAL EXPLOSON BEHAVIOR OF HYDROGEN/AIR MIXTURES IN OPEN SPACE

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#### ABSTRACT

In this study, the flame propagation behavior and the intensity of blast wave by an accidental explosion of a hydrogen/air mixture in an open space have been measured simultaneously by using soap bubble method. The results show that the flame in lean hydrogen/air mixtures propagated with a wrinkled flame by spontaneous instability. The flame in rich hydrogen/air mixtures propagated smoothly in the early stage, and was intensively wrinkled and accelerated in the later stage by different type of instability. The intensity of the blast wave of hydrogen/air mixtures is strongly affected by the acceleration of the flame propagation by these spontaneous flame disturbances.

## **1.0 INTRODUCTION**

Hydrogen is expected to serve as a clean carrier of energy, hence the considerable focus on basic technology and equipment relating to the utilization, manufacturing, transportation, storage and supply of this substance. For instance, successfully constructing a fuel-cell vehicle with hydrogen is remarkable. However, hydrogen has some serious properties in comparison with other combustible gas, for example, having an ignition energy as low as 0.019 mJ at stoichiomeric mixture, a wide flammability range in air (4-75% by volume of hydrogen), and its high diffusivity. If hydrogen leaks from hydrogen handling systems to the ambient air, there is a serious danger that the leaked hydrogen may be ignited easily by a very small energy, such as an electrostatic spark discharge, and this may lead to serious fire and/or explosion accidents. Therefore, the development of safety countermeasures based on hazard analysis and risk assessment assuming fire and/or explosion accidents is indispensable for the practical use of hydrogen in the social infrastructure. Especially, hazard analysis on an accidental explosion is very important because gas explosion causes indeed serious damages.

The main damages by accidental explosion are often caused by pressure increase, blast wave, fragment scattering, and so on. The damage caused by a blast wave can spread quickly and widely, and become a significant consequence of the accidental explosion around the explosion point. The type of blast wave which includes both sonic compression waves, shock waves and rarefaction waves, depends on how and when the energy is released in the explosion and the distance from the explosion area. The blast wave also depends on the flame propagation velocity and flame acceleration.

A lot of research results concerning the explosion of hydrogen-air mixture have reported. Especially, in order to evaluate the strength of the hydrogen/air mixture explosion, unconfined large scale experiments were recently carried out [1, 2, 3]. However, there has been little systematic research on the relation between flame propagation and blast wave in unconfined space. In this study, we conducted hydrogen-air deflagration experiment using soap bubble method to understand the relationship between flame propagation behaviour and blast wave in open space. We especially considered the effect of hydrogen/air mixture concentration to behaviour of flame propagation and blast wave.

#### 2.0 EXPERIMENTAL SETUP

The schematic diagram of the experiment setup is shown in Fig. 1. In order to measure simultaneously flame propagation behaviours and the blast wave in an open space, we used soap bubble method. The experimental apparatus consisted of the gas supplying system, the ignition system, the high speed schlieren photography system and sound pressure measuring system. A spherical soap bubble with hydrogen/air mixtures was made by premixed combustible gas flow nozzle. Ignition was produced at

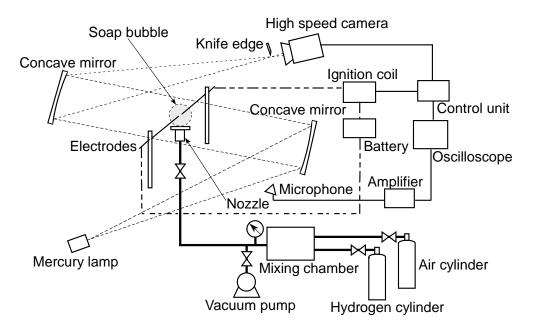


Figure 1. Experimental setup.

the center of the bubble by an electric spark generated by discharge electrode made of tungsten with a diameter of 0.5 mm and a high voltage ignition coil which transforms 12 volts (DC) to thousands of volts.

The flame propagation phenomena in an open space was imaged with schieren photography and recorded using a high speed camera (Photron, FASTCAM SA2) at 2000 frames per a second (exposure = 1/35000 s). The blast wave in an open space was simultaneously measured by microphone (PCB, 377B02, sensitivity = 50 mV/Pa), which in 0.3 m away from the spark gap. The pressure waveform obtained from the pressure sensors was amplified and recorded on a digital oscilloscope (Yokogawa, DL716).

In this study, the concentration of the gas mixture was determined by the partial pressure method and the equivalence ratio,  $\phi$ , was varied as follows: 0.7, 1.0, 1.3, 1.5, 1.8, 2.0, 2.5, 3.0, 3.5, and 4.0. A diameter of a spherical soap bubble with hydrogen /air mixtures was about 100 mm throughout the experiments.

### **3.0 RESULTS AND DISCUSSIONS**

#### **3.1 Flame propagation**

Figure 2(a) and (b) show detail of Schlieren picture. Figure 2(a) shows the picture before a combustible mixture ignites and Fig. 2(b) shows the picture when the flame is propagating in the mixture. It is shown that the electrodes are inserted into the bubble. It was different at each experiment whether the flame front was reached to the boundary between mixture and surrounding air simultaneously or previously when the bubble surface was ruptured. Figure 3 and 4 shows the schlieren images when the equivalence ratios are 0.7, 1.0, 1.8, 2.5, 3.0 and 4.0. When the equivalence ratio is 2.5 and under, in the early stage of flame propagation when the flame radius is small, an onset of stability in flame front is not observed. On the other hand, when the equivalence ratio is 3.0 and over, the wrinkles in flame front is not observed when the flame radius is small. Furthermore, the onset of the different type of instability in flame front was observed when flame front had reached to the non-

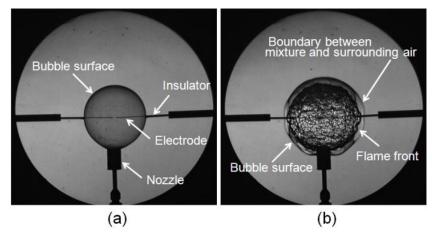


Figure 2. Detail of schlieren picture: (a) before ignition, (b) flame propagation process.

uniformity transition areas of concentration distribution of combustible gas. This transition area was formed by the rupture of soap bubble and mixing of combustible gas with surrounding air.

The flame radius versus time at various equivalence ratios was obtained from schlieren images. Figure 5 indicates that flame propagating velocity depends on the concentration distribution of hydrogen. Although the flame radius increases almost linearly with time in the early stage of flame propagation at  $\phi = 0.7$ , the flame propagation velocity gradually decreases in the later stage. On the contrary, in the case of  $\phi = 4.0$ , the flame propagation velocity rapidly accelerated in the later stage. In other case, the flame propagation velocity increases linearly or acceleratingly with time during the all stage. The acceleration of the flame propagation velocity is associated with the flame wrinkling.

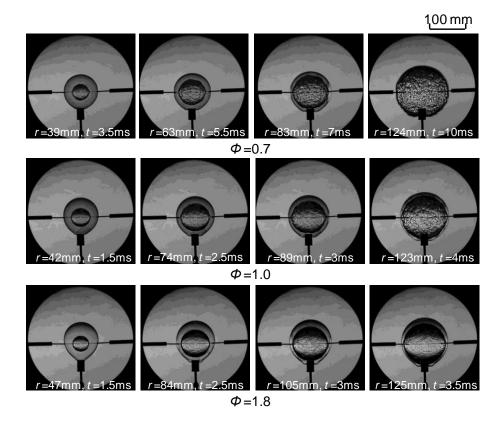


Figure 3. Schlieren images of flame propagation at equivalence ratios of 0.7, 1.0, 1.8.

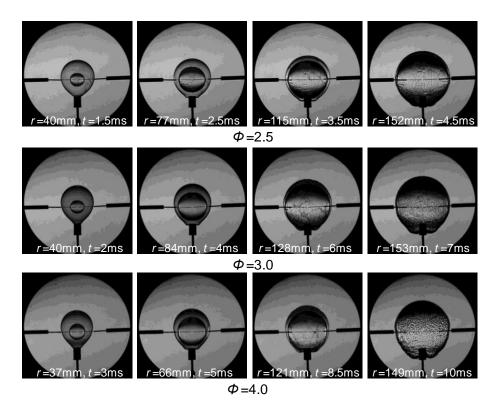


Figure 4. Schlieren images of flame propagation at equivalence ratios of 2.5, 3.0, 4.0.

Figure 6 shows comparison between measured mean burning velocity and literature data. Although the value of burning velocity is different from each experiment, the tendency is almost same. That is, the burning velocity is maximum at approximately  $\phi = 1.8$ . Present data are almost same as the data which were obtained by Aung et al [4].

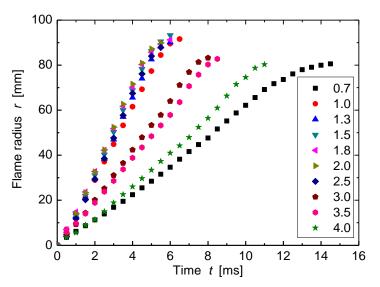


Figure 5. Flame radius versus time at various equivalence ratios.

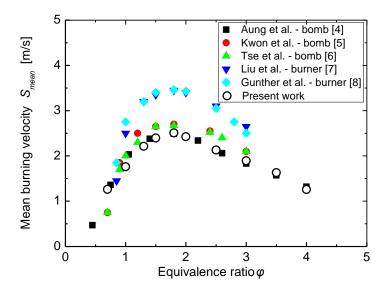


Figure 6. Comparison between measured mean burning velocity and literature data.

## 3.2 Blast wave

The blast wave in an open space was simultaneously measured by a microphone when the gas explosion occurred by the soap bubble system. The ignition spark and the time base of the oscilloscope were triggered simultaneously so that initial part of oscillogram corresponded to the time taken for the blast wave to propagate from the bubble to the microphone. Figure 7 shows the pressure wave histories for hydrogen-air bubble, of initial diameter 100 mm approximately at various equivalence ratios. The pressure in the early stage of flame propagation raises linearly with time at all equivalence ratios. It is shown that the measured overpressure increases more rapidly in the early stage as the burning velocity of the mixture becomes faster. The overpressure increases most rapidly in the

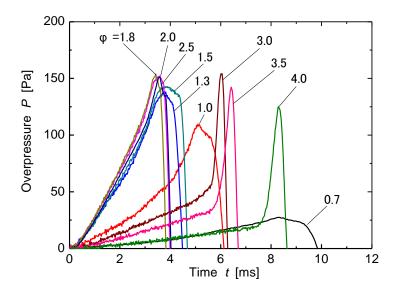


Figure 7. Pressure wave histories with different equivalence ratio.

mixture of  $\phi = 1.8$ . In more rich mixture,  $\phi = 3.0, 3.5, 4.0$ , the overpressure increases sharply in the last stage of flame propagation.

## 3.3 Comparison with exiting model by acoustic theory

If the expanding laminar front may act as a simple monopole source radiating spherically, it can be shown that at a distance, d, from the ignition position, the amplitude of the pressure fluctuation, p, where  $\rho$  is the density of the medium through which the sound is propagating as the sound wave is given by

$$p = \frac{\rho}{4\pi d} \frac{d}{dt} \left( \frac{dV}{dt} \right) \tag{1}$$

dV/dt is the rate of volume variation of the source [9]. Thomas et al. [10] proposed a theoretical expression of the pressure wave, which was observed as the flame propagates radially outwards at a constant velocity which equals to the product of burning velocity and volumetric expansion ratio. They affirmed that the pressure history can be expressed by the acoustic theory, and the peak positive pressure  $p_{max}$  of the blast wave can be expressed as the following equation.

$$p_{\max} = 2\frac{\rho}{d}\varepsilon(\varepsilon - 1)r_q S^2$$
<sup>(2)</sup>

Where  $r_q$  is the radius corresponding to the time at which peak overpressure was attained, S is burning velocity,  $\varepsilon$  is volumetric expansion ratio. This model shows that the peak overpressure depends on the volumetric expansion ratio and square of the burning velocity.

Figure 8 shows comparison between measured peak overpressure and predicted one calculated by Eq. (2) at various equivalence ratios. The burning velocity of existing literature [4] was used to calculate the overpressure at a constant burning velocity. When the equivalence ratio is 1.8 and under, the peak overpressure measured by experiments and evaluated value based on Eq. (2) show same tendency although measured data is larger than calculated data. In the case of  $\phi > 1.8$ , the equivalence ratio

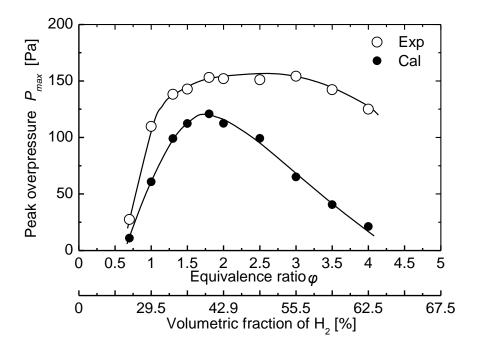


Figure 8 Comparison between measured peak overpressure and predicted one calculated by Eq. (2) at various equivalence ratios.

becomes high, the calculated data of the peak overpressure decreases gradually although the experimental data hardly decreases. It is shown that it is not appropriate for evaluating overpressure to use a mean burning velocity as the value of *S* on Eq. (2). In the rich hydrogen-air mixture, the flame wrinkling in later stage is generated when the flame approaches to the boundary between the mixture and surrounding air. Therefore, the rupture of soap bubble and heterogeneous mixing of the mixture with the surrounding air will be concerned. In the rich mixture, mixing with surrounding air will result in the increase of burning velocity. That is, the flame wrinkling in rich mixture might be generated by non-uniformity of concentration distribution of combustible gas. In this case, not only the burning velocity became a larger value, but also the burning velocity is acceleratingly increased. It means Eq. (2) can not apply to this case because Eq. (2) was derived under the assumption of constant burning velocity. Under the condition when the burning velocity is not constant, the following equation can be derived from Eq. (1).

$$p = \frac{\rho}{d} \frac{(\varepsilon - 1)}{\varepsilon} \left\{ 2\varepsilon^3 S^3 t + r^2 \frac{dS}{dt} \right\}$$
(3)

The second term in the right hand side of Eq. (3) indicates the contribution of the acceleration of burning velocity. To verify the Eq. (3), the time histories of flame radius, burning velocity (calculated from measured flame radius variation), and overpressure at equivalence ratios of 0.7, 1.8, 3.0 are plotted in Fig. 8. The flame radius in  $\phi = 0.7$ , 1.8 linearly increases with time. In comparison, in the case of  $\phi = 3.0$ , the flame radius linearly increases with time until about 5 ms, however, beyond 5 ms, the flame radius acceleratingly increased. It means that the second term in Eq. (3) has considerable effect only for the case of rich mixture.

Figure 9, 10 and 11 show that the overpressures predicted by Eq. (3) well agree with the measured histories of overpressure. The predictions can be realized by using the real burning velocity measured in the experiment. It is considered that the large peak overpressure in the later stage is caused by the acceleration of the burning velocity in the later stage. As a result, the overpressure of blast wave can be predicted by Eq. (3) derived from the acoustic theory if the real burning velocity could be known. The equation indicates that the intensity of blast wave is affected by burning velocity, volumetric expansion ratio and the flame acceleration. In particular, the intensity of the blast wave is strongly affected by the acceleration of the burning velocity.

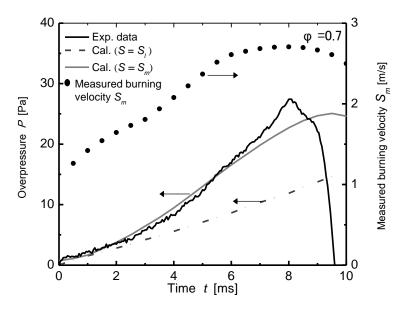


Figure 9. The time histories of flame radius, burning velocity, overpressure at equivalence ratio of 0.7.

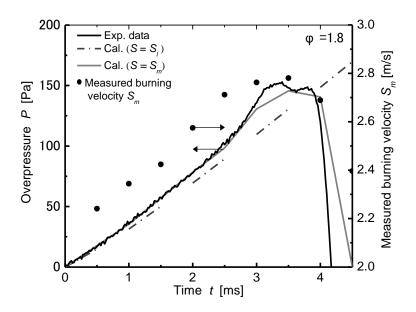


Figure 10. The time histories of flame radius, burning velocity, overpressure at equivalence ratio of 1.8.

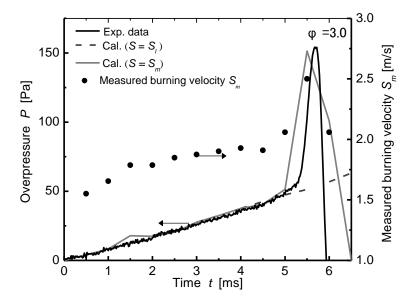


Figure 11. The time histories of flame radius, burning velocity, overpressure at equivalence ratio of 3.0.

uniformity formed during the mixing process of rich combustible mixture with surrounding air has a potential hazard of strong blast wave, because the mixing with air results in the increase of the burning velocity.

## **4.0 CONCLUSIONS**

Fundamental study on gas explosion in an open space has been carried out. The intensity of blast wave, which is of the highest risk, has been examined experimentally on the explosion of premixed hydrogen-air mixtures in an open space by using soap bubble method. Also the flame propagation behaviour has been measured. The main conclusions are summarized as follows;

1) The measurements of the intensities of blast wave show that; in lean hydrogen-air mixture the overpressure grew linearly with time, in rich hydrogen-air mixture the overpressure grew linearly with time in the early stage and acceleratingly increase in later stage. The accelerating increase in the later stage resulted in a much larger peak overpressure than that in the stoichiometric mixture.

2) The overpressure of blast wave can be predicted by the acoustic theory if the real burning velocity could be known. The theory indicates that the intensity of blast wave is affected by burning velocity, volumetric expansion ratio and flame acceleration. In particular, the intensity of the blast wave is strongly affected by the acceleration of the burning velocity.

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