**Explosion characteristics of hydrogen–nitrogen–air mixtures at elevated pressures and temperatures**

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**ABSTRACT**

An experimental study on the combustion characteristics of nitrogen diluted hydrogen was conducted in a constant volume combustion vessel over a wide range of equivalence ratios and dilution ratios at elevated pressures and temperatures. The explosion characteristics such as the explosion pressure, the combustion duration, the maximum rate of pressure rise, the deflagration index and the normalized mass burning rate were derived. The result shows that a short combustion duration and higher normalized mass burning rate were presented with the increase of equivalence ratio. With the increase of initial temperature, the explosion pressure, the maximum rate of pressure rise and the deflagration index were decreased, and a shorter combustion duration and higher normalized mass burning rate were presented. With the increase of initial pressure, the explosion pressure, the maximum rate of pressure rise and the deflagration index increase, a shorter combustion duration and higher normalized mass burning rate were presented. Nitrogen dilution significantly reduces the normalized mass burning rate and the deflagration index and thus the potential of explosion hazards.

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**1. Introduction**

Limited fossil fuel availability and continuously stringent restrictions on engine pollutants have intensified the study on alternative fuels around the world. Hydrogen is regarded as one of the most promising fuels and the use of hydrogen in various propulsion and energy applications, such as in fuel cells and internal combustion engines is being extensively studied. The wide flammability range of hydrogen enhances the lean burn capacity which would result in a low NOx emission level [1–3]. The high burning velocity of hydrogen facilitates the constant volume combustion at top dead center and this may contribute to a relatively higher thermal efficiency. In addition, the low minimum ignition energy of initiation of hydrogen flame kernel could reduce the cycle by cycle variations [4,5]. However, one of the major problems associated with applying hydrogen is the combustion-induced disasters such as fires and explosions [6–11]. The damages in fires are mainly caused by high heat flux from the flames and the toxic smoke from combustion [12]. Explosion in confined spaces is a well recognized hazard due to the high explosion pressure and high rate of pressure rise. This is because the fast heat release during combustion could not be dissipated by slow process of the heat conduction, convection, and/or radiation before the blast wave reached the sidewall of the confined space [13]. Fire is the most common incidents in the

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process industry. However, the proportion of losses due to fire is only 20%, while explosions contribute a remarkable 75% of the total losses, and the rest belongs to the toxic release [14,15].

The explosion pressure, $P_{max}$ and the maximum rate of pressure rise $(dP/dt)_{max}$ are the most important explosion severity parameters in assessing the hazards of a process, for design of vessels to tolerate explosion pressure and for design of relief device against damage from gaseous explosion [16]. The explosion time ($t_c$) is the time interval between ignition and the moment when $P_{max}$ is attained. It is noted that the explosion time, $t_c$, is sometimes defined as the combustion duration [14,17,18]. Because $(dP/dt)_{max}$ depends on the size of the vessel, it is often normalized with respect to the volume of the vessel according to the cubic-root law [16,19-21],

$$\left( \frac{dP}{dt} \right)_{max} V^{1/3} = K_c$$

Here, $K_c$ is an explosion severity factor called deflagration index. The deflagration index is an indication of the 'robustness' of the explosion. It was assumed that $K_c$ is a constant regardless of the vessel volume, and depends only on the mixture composition [21]. Therefore, for practical purpose, $K_c$ of gas mixtures at standard temperature and pressure facilitates scaling the explosions in such vessels. As shown in Table 1, the higher the deflagration index is, the more robust the explosion and the greater the consequences of the explosion will be [11,21]. $P_{max}$, $(dP/dt)_{max}$, $t_c$, and deflagration index $K_c$, are influenced by initial temperature and pressure, initial or combustion generated turbulence, dilution and fuel-air ratio [16,19,22]. The addition of diluents can obviously mitigate the explosion severity due to the increased specific heat of non-fuel gases per unit oxygen concentration [23]. Theoretically, for given mixture composition and initial pressure and temperature, the maximum value of explosion pressure is the adiabatic equilibrium pressure ($P_e$). Because of the heat loss caused by thermal conduction, convection, or radiation, the measured maximum pressure ($P_{max}$) of an explosion in closed vessel is always lower than the adiabatic equilibrium pressure [24].

The rate of combustion is essential for pressure evolution in closed vessel explosions [20]. This is because the maximum pressure, the maximum rate of pressure rise, the deflagration index and the combustion duration is dependent on the amount of mass burning and the rate of mass burning which determines the amount and the rate of heat release. At elevated temperatures and pressures, the mass burning rate is always a function of initial charge density, which depends on the pressure and temperature. It is then essential to define a normalized mass burning rate (or heat release rate) to exclude the influence of initial charge density on mass burning rate, especially at elevated pressures and temperatures. Lewis and von Elbe [25] gave an expression which related the burned mixture mass to the fractional pressure rise. Based on this equation, the burned mass fraction can be expressed as,

$$m_b = \frac{P - P_u}{P_e - P_u} m_0$$

where $m_b$ and $m_0$ are the burned mass and initial mass of the charge. $P_u$, $P_e$ and $P$ are the adiabatic equilibrium pressure, the initial pressure and the instant pressure, respectively. By differentiating Eq. (2) with respect to time, the normalized mass burning rate is obtained,

$$\frac{1}{m_0} \frac{dm_b}{dt} = \frac{1}{P_e - P_u} \frac{dP}{dt}$$

In this work, explosion characteristics such as explosion pressure, maximum rate of pressure rise, combustion duration, deflagration index and the normalized mass burning rate are obtained to evaluate the hazards of hydrogen explosion in the presence of nitrogen dilution at elevated pressures and temperatures.

### 2. Experimental setup and procedures

In this work, the dilution ratio is defined as the volumetric fraction of nitrogen addition in the pre-mixtures:

$$\phi_i = \frac{V_{diluent}}{V_{H_2} + V_{air} + V_{diluent}}$$

As shown in Fig. 1, the experimental apparatus consists of the mixture preparation manifold, the combustion vessel, the heating system and the data acquisition system. Fig. 2 shows the schematic diagram of the cylinder-type combustion vessel with diameter of 180 mm and length of 210 mm. Two sides of the vessel are mounted with the quartz windows to allow the optical access. The combustion vessel is designed to withstand a maximum internal pressure of 10 MPa. A pressure transducer (Kistler 7001) at a sample-rate of 100 kHz, connected with a Charge Amplifier (Kistler 5011) was used to record the dynamic pressure during explosions. The mixtures were prepared by introducing each component according to its corresponding partial pressure at the specified equivalence ratio. An external electric heating circuit is incorporated at the outside surface of the combustion vessel to facilitate uniform heating of the mixture within the vessel to a desired initial temperature. The mixtures are ignited by the centrally located electrodes. A standard capacitive discharge ignition system is used to produce the spark. Once the combustion was completed, the combustion vessel was vacuumed and flushed with dry air three times to avoid the influence of the residual gas on the next experiment. A time interval of 5 min was awaited to allow the mixtures to be quiescent and to avoid the influence from the wall. At high temperatures and pressures, time interval of 30 min was tested for several mixtures, and no appreciable difference in pressure history for given mixture composition was observed compared to the time interval of 5 min. This ensures the accuracy in filling process and no
mixture leakage. Purities of nitrogen and hydrogen in the study are 99.96% and 99.99%, respectively.

Combustion pressure evolution of various equivalence ratios and dilution ratios at elevated pressures and temperatures was measured. The adiabatic equilibrium pressure was obtained by the Equilibrium model with CHEMKIN. As shown in Fig. 3, the experimental pressure–time curve contains pressure oscillation, so it is necessary to determine the mean pressure–time curve and its derivative by some smoothing filter. In this work, the Savitsky–Golay method [26] is adopted as suggested by Razus et al. [16] and Dahoe [27]. This was done with an Origin 7.0 by applying a second order polynomial and 21 points (a 10 left and 10 right) data window. It is seen from Fig. 3 that the points are very heavily scattered and the determination of maximum rate of pressure rise would be meaningless if the smoothing filter is not introduced. In this work, the combustion pressure evolution is filtered in this way before further processing. As observed in the experiments, all pressure time curves exhibit the similar behavior: after ignition, the pressure increases progressively until the maximum value of pressure rise rate \((dP/dt)_{\text{max}}\) is achieved, then continues to increase with a decreasing pressure rise rate towards the explosion pressure, \(P_{\text{max}}\), and after that, the pressure begins to decrease. This behavior is a typical pressure evolution process in a closed vessel, similar to the observations by Dahoe for methane-air flame [28] and hydrogen-air flame [27].

3. Results and discussions

Fig. 4 illustrates the pressure evolution during combustion process. The pressures are normalized with respect to the initial pressure \((P_u)\). Fig. 4(a) gives the pressure–time curves at different equivalence ratios. The figure shows that all pressure–time curves have a similar shape and both lean and rich mixture give a lower value of explosion pressure \(P_{\text{max}}\), compared with that of the stoichiometric condition. Meanwhile, the phasing at \(P_{\text{max}}\) is advanced with the increase of equivalence ratio. At the equivalence ratio of 1.0, the adiabatic
flame temperature is higher than that of the off-stoichiometric mixture due to larger heat release and this leads to the highest explosion pressure at the equivalence ratio of 1.0. In addition, the flame propagation speed is increased with the increase of equivalence ratio, and thus the timing at $P_{\text{max}}$ is advanced. Fig. 4(b) illustrates the influence of initial temperature ($T_u$) on the pressure evolution. With the increase of $T_u$, $P_{\text{max}}$ is decreased and the timing at $P_{\text{max}}$ is shortened. The amount of unburned mixture is decreased and hence the heat release is decreased with the increase of $T_u$. The increase in $T_u$ will increase the adiabatic temperature and hence the flame propagation speed. This results in the faster pressure rise with the increase of $T_u$. Fig. 4(c) gives the pressure evolution process at different initial pressures. $P_{\text{max}}$ is increased significantly with the increase of $P_u$. However, the maximum peak value of normalized pressure is moderately sensitive to the increase in $P_u$. In addition, increasing $P_u$ results in faster pressure rise. Fig. 4(d) clearly shows that the effect of nitrogen suppression on the explosion pressure and the rate of pressure rise. The maximum peak pressure is decreased significantly and the pressure rise becomes slower with increasing dilution ratio ($\phi_r$). Obviously, the total amount of heat release decreased with the increase of dilution ratio. The specific heat of the non-fuel gas per unit oxygen concentration increases and hence the flame propagation speed is decreased as nitrogen is added [23], and these result a lower pressure rise with increasing dilution ratio.

Fig. 5 gives the normalized peak pressure ($P_{\text{max}}/P_u$) and the combustion duration ($t_c$) versus equivalence ratio. It is clear that $P_{\text{max}}/P_u$ gives its maximum value at stoichiometric mixture condition, regardless of the initial temperature, initial pressure, or dilution ratio. Since the adiabatic temperature is directly controlled by the heat release, the behavior that $P_{\text{max}}/P_u$ gives its maximum value at the equivalence ratio of 1.0...
reflects the dominant effect of adiabatic temperature. The combustion duration, $t_c$, decreases monotonically with the increase of equivalence ratio. This reveals the dominant effect of flame propagation speed on the pressure evolution process. Actually, for almost all kinds of fuels, the adiabatic temperature and the flame propagation speed give their maximum values at the stoichiometric mixture condition. However, due to the highly diffusive nature of hydrogen [29], the peak value of flame propagation speed well shifts to the fuel rich side and this shortens the combustion duration with increasing the equivalence ratio. In Fig. 5(a), $P_{\text{max}}/P_u$ is decreased with the increase of initial temperature due to the decreased mass of the charge and hence the combustion heat release. $t_c$ is decreased with the increase of initial temperature due to the increased flame propagation speed. In Fig. 5(b), $P_{\text{max}}/P_u$ is increased with the increase of initial pressure. At higher initial pressures, the flame tends to be turbulent more easily because of the promoted flame front instability with increasing the pressure. This increases the flame propagation speed [22] and shortens the combustion duration. Fig. 5(c) shows the effect of nitrogen dilution on the explosion pressure and the
combustion duration. Nitrogen addition can significantly reduce the explosion severity. The adiabatic flame temperature and the flame propagation speed are decreased because of the increased specific heat and the decreased amount of heat release.

Fig. 6 gives the maximum rate of pressure rise and the deflagration index versus equivalence ratio at different initial temperatures, pressures and dilution ratios. For most cases, the deflagration index is higher than 30 MPa m s⁻¹ (St 3), indicating that hydrogen is of high explosion hazard potential. In addition, both \( \frac{dP}{dt} \)max and \( K_G \) in the concerned equivalence ratio range will be increased with the increase of equivalence ratio.

As shown in Fig. 6(a), \( \frac{dP}{dt} \)max and \( K_G \) are decreased with the increase of initial temperature. Increasing \( T_u \) will increase the flame propagation speed and brings a faster pressure evolution process as shown in Fig. 4(b). However, the maximum rate of pressure rise and deflagration index are decreased with the increase of \( T_u \). Fig. 6(b) shows that the maximum rate of pressure rise and the deflagration index are increased dramatically with the increase in the initial pressure. With the increase of \( P_u \), the flame front becomes more unstable, the flame front will easily be developed into a turbulent type and increases the flame front area [12], resulting in a faster flame propagation speed and hence a higher rate of pressure rise [22].

Fig. 6(c) gives the maximum rate of pressure rise and deflagration index at different dilution ratios. The presence of increased nitrogen leads to a progressive decrease of fuel content and the heat release and the increase in specific heat. These will contribute to the decrease of \( \frac{dP}{dt} \)max and \( K_G \). At the dilution ratio of 0.1, the deflagration index is reduced to less than 30 MPa m s⁻¹, indicating that the nitrogen dilution can significantly degrade the deflagration index level and hence reduce the potential of explosion hazards.

Fig. 7(a) gives the instant normalized mass burning rate (NMBR) at different equivalence ratios. Initially, NMBR increases progressively until a maximum value is attained because of the increased flame front surface area. After the flame ball reaches the vessel wall, NMBR decreases due to decreasing of flame surface area. The maximum peak value of NMBR is attained at the stoichiometric mixture condition. NMBR increases more rapidly with the increase of equivalence ratio. Fig. 7(b) illustrates NMBR at different initial temperatures. NMBR increases faster with the increase of \( T_u \) because of the increased flame speed. Increasing temperature decreases the initial charge density, the maximum rate of pressure rise and deflagration index as shown in Fig. 6(a), and this may lead to the paradox that the explosion ‘robustness’ is decreased with increasing \( T_u \). However, by introducing the definition of NMBR, the influence of temperature on the charge density was excluded, and it shows that the peak value of NMBR is increased with the increase of \( T_u \). When the initial temperature is high, the heat transfer to the vessel wall is
decreased due to the decreased combustion duration and higher vessel wall temperature. Fig. 7(c) gives the NMBR at different initial pressures. At higher initial pressures, the flames are easily developed into turbulent type because of the promoted flame front instability [12]. This results in a faster increase of NMBR with increasing the initial pressure. The peak value of NMBR is increased with the increase of \( P_i \). Fig. 7(d) shows that the suppression effect of nitrogen addition on the normalized mass burning rate. Significant decrease in the peak NMBR is observed with the increase of dilution ratio. The timing at peak NMBR is postponed with the increase of dilution ratio due to the decreased flame propagation speed.

### References


