EFFECT OF HYDROGEN CONCENTRATION ON THE EXPLOSION SEVERITY IN A CLOSED VESSEL

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"I hereby declare that I has read this thesis and in my
opinion this thesis is sufficient in terms of scope and quality for the
award of the Degree of Bachelor Hons. (Chemical Engineering)."

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A thesis submitted in fulfilment of the
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DECEMBER 2016

I declare that this thesis entitled *"effect of hydrogen concentration on the explosion severity in a closed vessel"* is the result of my own research except as cited in the references. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

Signature : …………………………….....

Name : Mohd Aidil Bin Awaluddin

Date : …………………………….

I dedicated this thesis to my supervisor, Ir. Dr. Siti Zubaidah Binti Sulaiman, my lovely parents, teammates, and friends. I couldn’t go to this far without you. Thank to you to all your support during my Degree journey.

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In the name of Allah, The Most Gracious, The Most Merciful. Praise is to Allah S.W.T. by whose grace and blessings I receive guidance in completing this study. Thanks for His greatest love and blessing. May Allah S.W.T. also bless The Prophet Nabi Muhammad S.A.W. and grant him and his family peace.

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ABSTRACT

Hydrogen gas can be produced from steam reforming natural gas, and steam gasification process. Hydrogen is widely used for producing ammonia for fertilizer, petroleum refining, glass purification and many others application. Through the rapid discoveries for the hydrogen production, this lead to the severity of hydrogen explosion phenomenon and tough safety precautions in term of quantitative and qualitative were required. Thus, this study will discover the severity limit of hydrogen explosion. The objectives in this study are to know the maximum rate of pressure rise $(dp/dt)_{max}$, maximum explosion pressure (P_{max}), and to study the explosion in different vessel shapes. Pure hydrogen (99.9%) was used as the fuel and mix with air. The experiment were conducted in a 20-L spherical bomb with hydrogen content in air at 30% v/v and different equivalence ratio from Ø = 0.4 to 1 in Combustion Laboratory, Faculty of Chemical Engineering, Universiti Malaysia Pahang with two igniters of 10kJ at the centre of the vessel. The experiment was repeated three times to get the fit result. From the result obtained the maximum explosion pressure, P_{max} and rate of pressure rise, dP/dt increase with increase of hydrogen concentration in air. This is due to the increase amount of fuel and more chemical reaction happened during the explosion. Higher explosion parameters were happened at stoichiometry concentration or at equivalence ratio 1 with P_{max} 5.4 bar and dP/dt 1410.3 bar/s. In comparison between spherical and 2-in pipe and 4-in pipe, the 2-in pipe has higher explosion pressure of 9.1 bar at equivalence ratio 1. This is due to the quenching effect in larger diameter (0.1 m in 4-in pipe and 0.34 m in spherical vessel) plays a significant role on the explosion pressure development. Different result obtained on rate of pressure rise, dP/dt in these three vessel shapes. Explosion in the spherical vessel tends to have higher value of dP/dt at 1410.3 bar/s. High dP/dt means amount of burning rate and pressure generation to be released from the explosion is has severe condition. Thus this concluded that hydrogen-air mixture explosion is more severe in spherical vessel compare to explosion in cylindrical vessel.

ABSTRAK

Gas hidrogen dihasilkan daripada proses wap pembaharuan gas asli, dan proses pengegasan wap. Hidrogen digunakan di dalam proses penghasilan gas ammonia untuk penghasilan baja, penapisan petroleum, pengaslian gelas dan pelbagai proses yang lain. Akibat daripada penghasilan gas hidrogen yang meningkat, keadaan ini mendorong kepada risiko bahaya terhadap letupan gas hydrogen di industry dan tahap keselamatan yang ketat dari segi kualitatif dan kuantitatif diperlukan. Oleh itu, kajian ini dijalankan untuk mengkaji tahap kebahayaan letupan gas hydrogen. Objektif kajian ini ialah untuk mengetahui kadar peningkatan tekanan maksima $(dP/dt)_{max}$, tekanan letupan maksima (P_{max}), dan mengenali parameter letupan di dalam bentuk sfera dan bentuk silinder. Gas hydrogen asli (99.9%) digunakan sebagai bahan bakar dan dicampurkan dengan udara. Eksperiment ini menggunakan bom sfera bersaiz 20-L yang mengandungi dua penyala berkuasa 10kJ dengan kandungan hydrogen di dalam udara ialah 30 v/v% dan pada nisbah hydrogen-udara yang berlainan iaitu Ø = 0.4 sehingga 1 yang terdapat di dalam Makmal Kejuruteraan Kimia dan Sumber Asli (FKKSA) di Universiti Malaysia Pahang. Eksperimen ini diulang sebanyak tiga kali untuk mendapatkan hasil bacaan yang konsisten. Daripada keputusan yang dihasilkan, iannya mendapati bahawa tekanan letupan maksima (P_{max}) dan kadar peningkatan tekanan maksima $(dP/dt)_{max}$ meningkat dengan peningkatan nisbah hydrogen di dalam udara. Ini disebabkan oleh peningkatan jumlah bahan bakar dan reaksi kimia yang berlaku sewaktu letupan. Parameter letupan yang kuat berlaku pada kepekatan stoikiometri 1 dengan P_{max} 5.4 bar dan dP/dt 1410.3 bar/saat. Perbandingan diantara bentuk sfera dan paip pula mendapati, tekanan letupan maksima (P_{max}) di dalam paip 2-in lebih kuat pada 9.1 bar berbanding paip 4-in dan sfera. Ini disebabkan oleh kesan pelindapkejutan di dalam paip yang mempunyai saiz diameter besar (0.1 m di dalam paip 4-in dan 0.34 m di dalam sfera) memainkan peranan signifikan terhadap tekanan letupan yang terjadi. Kadar peningkatan tekanan maksima $(dP/dt)_{max}$ pula tinggi di dalam bentuk sfera dan ia menunjukkan letupan gas hydrogen-udara di dalam sfera adalah jauh lebih bahaya berbanding letupan di dalam paip silinder.

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DDT	Deflagration Detonation Transition
dP/dt max	Maximum rate of pressure rise
LFL	Lower Flammability Limits
LPG	Liquefied Petroleum Gas
LOC	Limiting Oxygen Concentration
P_{max}	Maximum rate of explosion pressure
S_f	Flame speed
S_g	Unburned gas velocity
S_L	Laminar flame speed
UFL	Upper Flammability Limits
W	Molecular Weight
ρ	Density
φ	Equivalence ratio

CHAPTER 1

INTRODUCTION

1.1 Background

Research and innovations of the prevention of gas explosion is always been developed by using advance technologies. The hydrogen gas production is continuously processed as the demand for its' usage growing rapidly. Hydrogen gas can be produced from steam reforming natural gas, and steam gasification process. It is widely used for producing ammonia for fertilizer, petroleum refining, glass purification and many others application. According to Fuel Cell & Hydrogen Energy Association's Fact Sheet, another uses of hydrogen is as a clean fuel for fuel cell electric vehicles (FCEVs).

In most industries, the hydrogen gas was transferred from one section to another section by using pipelines and hydrogen storage in a circular tank. However, one of the major problems occurred when applying hydrogen usage is the combustion and explosion accidents in industries (C. Tang et al., 2009). A Neville (2009) stated that an explosion cannot occur in a tank or closed vessel contains only hydrogen. Thus, an oxidizer such as oxygen and source of ignition must be present to start the explosion with hydrogen concentrations content range from 18.3% to 59% equivalence to flammability range from 4% to 74% in air (Xueling Liu, and Qi Zhang, 2014).

In the summer 1985 in Norway, a severe hydrogen-air explosion occurred in an ammonia plant. Bjerketvedt, D and Mjaavatten, A (2005) reported that the incident is one of the largest gas explosions in industrial hydrogen explosion. Three men were seriously injured and destruction of the building of the explosion occurred were resulted from the explosion. The source of the explosion was happened at pipe leakage at one of the operating pump that feeding water to a vessel containing hydrogen at a pressure of 30 bars. The pressure push the water flow to back flow and reach the leaking point. Hydrogen discharge at the leakage point lasted about 20 to 30 seconds before the explosion occur which also released 10 to 20 kg hydrogen gas. The incident causes a large horizontal jet flame for about 30 seconds.

Another hydrogen-air explosion incident occurred at China Light and Power Cast Peak Generating Station in August 28, 1992. The explosion happened in confined vessels which are two receivers that supply hydrogen to a generator. The plant was shut down on August 24 to 26 and resume to supply hydrogen to receivers on August 27. Suddenly the hydrogen purity in the generator dropped to 85% and the receivers were disconnected from generator for hydrogen purity sampling. The sampling shows the purity is 95% and then the two receivers were reconnected to supply hydrogen to generator. Again, the purity in the generator indicated 85% hydrogen purity. 20 minutes later, both receivers exploded at 10:05 a.m. which resulting two fatalities; 18 injured by fragments and the extensive blast damage in 100 m radius. An investigation from the incident reported that all the gas supplied to the receiver over 20 hour period was air. When hydrogen mixed with air at right concentration at 18.3% to 59%, explosion can be happen and this explained the cause of the explosion in the receivers.

Explosions are destructive phenomenon that could affect the economic implications and major social (M. Prodan et al, 2012). Therefore, it is necessary to know the explosion parameters and characteristic. A triangle explosion diagram simply explains the way on how an explosion occurred by three factors. The factors are oxygen concentration, fuel concentration, and source of ignition.

1.2 Motivation

Hydrogen gas usage, processing, and storing had been widely appeared in whole word. Hydrogen is a promising energy in the future and the demand of hydrogen gas will increases in the future. H. Xiao (2010) take the USA as an example, it was estimated that by 2040 the annual demand for hydrogen will reach 15 million tons. Rapid processing, transporting, and selling the hydrogen gas would result in applying for better technology to achieve the demand. But, lack information about hydrogen safety handling is the main reasons that initiate to the explosion. Thus this situation leads to the risk towards hydrogen explosion that could cause a serious accident in industries when no safety considerations are takes into account.

The safety practices in production, storage, distribution and use of hydrogen are key issues to hydrogen energy industrialization. In addition, this study can provide a basis for the development of preventive and control measures for explosion accidents of hydrogen and further study of gas activity.

1.3 Problem statement

Hydrogen explosion in air determines the explosion severity when explosion occurred in a vessel. When an explosion happened, the mass and energy content within the mixture mostly dominated by hydrogen as fuel are high enough to cause a massive explosion. Besides that, the explosion mechanism such as diffusivity, and flame structure encourage the explosion to reach the maximum parameter. Then, the resulting effect from the explosion is the rate of pressure rise and deflagration index which are two important measurements of explosion severity. Explosion characteristics study had been conducted through many researchers in various type of vessel shapes and dimensions.

C. Tang (2009), studied the explosion parameters such as maximum explosion pressure, P_{max}, maximum rate of pressure rise, $(dP/dt)_{max}$, deflagration index, K_G of hydrogen/nitrogen/air in a cylinder vessel. They found that the explosion characteristic above together with combustion duration are decreased, while normalized mass burning rate is increased with the increase of initial temperature. As the initial temperature increases, the amount of unburned mixtures decreased. Besides that, they found the explosion characteristic and normalized mass burning rate are increased, while the combustion duration is decreased with the increase of initial pressure.

M. Fagieh (2016) studied a computational study to know the deflagration index in hydrogen, methane, and their mixture. Differ from (C. Tang et. al., 2009), the vessel shape used in their experiment is a spherical vessel. Compare the explosion in a cylindrical vessel to a spherical vessel is the amount of unburned mixture in cylindrical vessel cannot be completely consumed by the propagating spherical flame as it reaches the inner wall of the vessel. This explosion phenomenon could effects the explosion characteristics by lowering the rate of pressure rise, $(dP/dt)_{max}$ and deflagration index, K_G. That is the reason why a spherical vessel should be used to measure the deflagration index. The result obtained from the experiment were compared to the other researchers (Holtappels, 2002; Jo and Crowl, 2010; Ma et al., 2014) result and found that there are substantial discrepancies in the deflagration index measurement. The discrepancies are might be caused by flame instability which accelerates flames propagation and thus, increases the maximum pressure rise rate and deflagration index.

Z. Y. Sun (2016) performed an experiment to studied laminar spherical flames within homogenous hydrogen-air mixture in a spherical vessel. Their outcome results were more on the global stretch effects on flame front for lean, stoichiometric, and rich, and Markstein length. On the other hand, (J. Guo, 2015), studied ignition point in vented vessel. J. Goulier et al (2016) studied the laminar and turbulent flame speed of a spherical flame in a fan stirred closed vessel for hydrogen safety application. They found that the evolution of the pressure inside the spherical vessel is strongly affected by the presence of the initial turbulence and the time for amount of fresh gases combusted is much fast.

Those researchers above were mainly discussed and found the explosion characteristic in both spherical and cylindrical vessel but there are still needed to in-depth understandings on the explosion characteristic at lean side towards the stoichiometry, and severity of explosion in type of vessel shapes. The factors contributing to the explosion such as diffusivity, mass burning rate, and flame structure that could affected the maximum explosion pressure, and rate of pressure rise need to have more discussion to give a clear figure of explosion severity for safety propose. Thus, it is need to know the understanding on the mechanism of the hydrogen-air mixture explosion by study the effect of equivalence ratio form lean to stoichiometry in both spherical and cylindrical vessel shape.

1.4 Objectives

The objectives of this research are:

i. To evaluate the effect of Equivalence Ratio (ER) of hydrogen – air mixture in explosion pressure and rate of pressure rise.
ii. To study the effect of vessel shape on the explosion pressure and rate of pressure rise.

1.5 Scope of research

The scopes of research in the study are:

i. The experiment will be conduct in a 20 L spherical bomb at the ambient condition pressure of 1 bar.
ii. The research will use the range of hydrogen concentration that is varied from equivalence ratio of 0.4, 0.6, 0.8 1.0.
iii. The research is conducted to find the maximum pressure, and rate of pressure rise.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

In hydrogen safety community, hydrogen explosion hazard is extremely important and has been extensively focused due to its wide flammable range. The wide flammability range of hydrogen enhances the lean burn capacity in explosion (F. Ma et al., T. Shudo et al., 2008, J.W. Heffel, 2003).

Other than hydrogen gas explosion, hydrocarbon gases such as methane, propane also could cause an explosion at right concentration with air. However, experimental researchers report a number of common findings, including the fact that when an explosion uses hydrogen fuels, Deflagration to Detonation Transition, DDT has the potential to be achieved at a magnitude of greater severity, compared to hydrocarbon fuels (Heidari and Wen, 2014; Thomas et al., 2010).

The high burning velocity of hydrogen facilitates the constant volume combustion at top dead centre 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 (F. Ma et al., J. Wang et al., 2008).

However, one of the major problems associated with applying hydrogen is the combustion-induced disasters such as fires and explosions.

2.2 Gas explosion categories

Figure 2.1 below shows the schematic diagram of detonation deflagration which occurs in an explosion. The explosion development can be categorizes into two types that are deflagration, detonation or may transit from deflagration to detonation during the explosion development.

Deflagration was defined as when the combustion wave propagates at a speed lower than the speed of sound. Different case when the combustion wave propagates at faster than the speed of sound (R. Blanchard et al., 2011). The temperature of the explosion is much higher than room temperature. As a result, the speed of sound increase with the increase of temperature, the explosion sound travelled very high. In this phenomenon when the combustion wave propagates more than the speed of sound at specific temperature is called detonation (Heidari and Wen, 2014). The pressure rise in detonation is much higher than deflagration. Compare to original pressure or initial pressure, the pressure may rise up to eight times in deflagration. On the other hand, the peak pressure may reach twenty times or more in detonation and the shock wave generated from in detonation are very ruinous.

Flame explosion within the limits of deflagration and detonation is called quasi-detonation. It happened when a transition occurs for a low speed deflagration flame. The phenomenon of the transformation of a low speed deflagrated flame to a catastrophic detonation explosion is called as Deflagration to Detonation Transition or DDT. The Deflagration to Detonation could happen with two factors that presence in the phenomenon which are obstacles and particular geometries of explosion galleries.

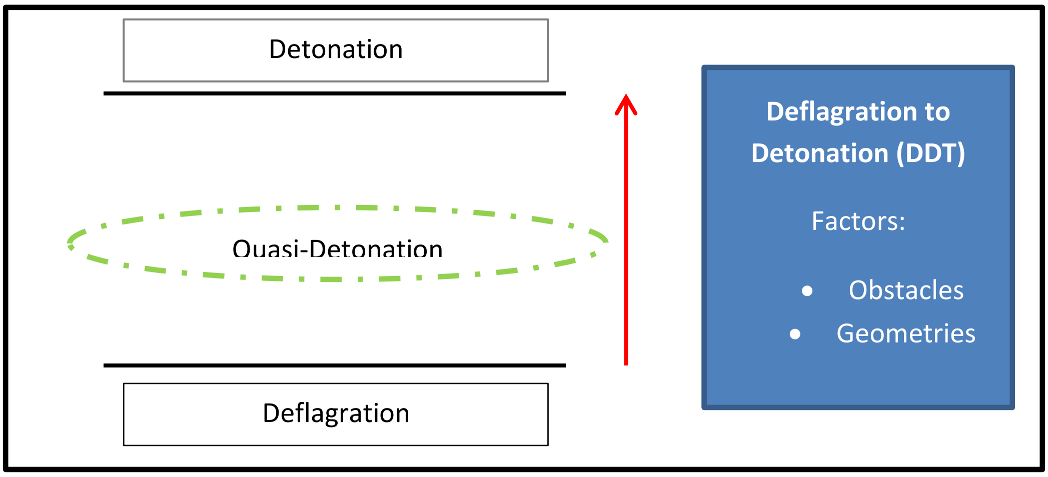

Figure 2.1: Schematic diagram of Deflagration Detonation.

2.3 Factor contributes to the explosion

Explosion in a closed vessel is a condition on which a mixture of flammable gas with air is burning with the flame propagation at specific concentrations. Razus et al. (2006) have studied the explosion pressures of hydrocarbon-air mixtures in closed vessels. They showed that the initial pressure on flammability limit, fuel concentration and type of vessel shapes have a significant effect on the maximum overpressure during flame propagation.

2.3.1 Initial pressure and temperature on flammability limit

Initial pressure and temperature have a significance correlation in a flammability limit. In a fire triangle explosion in figure 2.2, the concentration factors which are oxygen, fuel, and ignition source play an importance role to determining the explosion limits of the mixture. There are two types of flammability limits that are Upper Flammable Limit (UFL) and Lower Flammable Limit (LFL). The size of the limits that resulted from the three factors concentration will show the severity of the explosion. The bigger the size of the flammable limit, the more harmful of the explosion and vice versa. The size can be reduce off by introduced an inert into the mixture because of the chemical properties of the inert itself is non-react to the fire explosion.

H. J. Liaw (2009) briefly explained the properties of UFL and LFL as the upper flammability limit (UFL) is taken as the fuel molar concentration of a non-flammable mixture for which a 0.4 mol% leaner mixture is flammable, while the lower flammability limit (LFL) is taken as the fuel molar concentration of a non-flammable mixture for which a 0.2 mol% richer mixture is flammable.

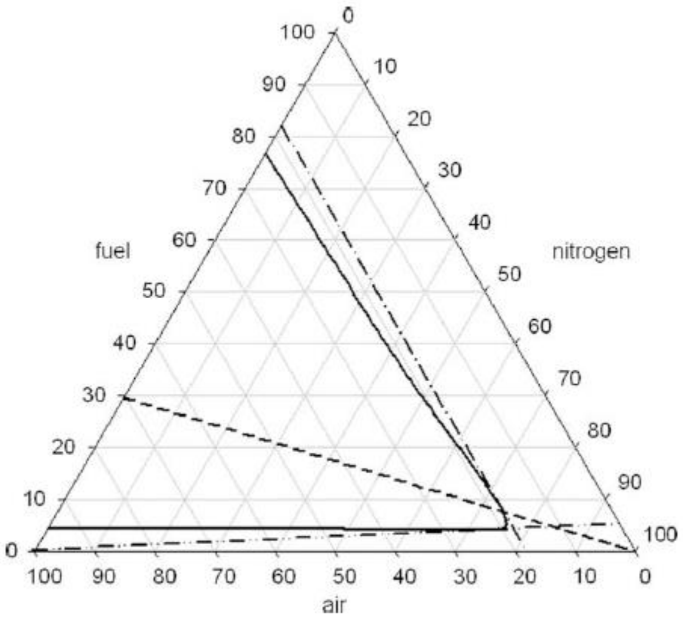

Figure 2.2: Fire triangle diagram that show the flammable range (solid line) of a fuel/air mixture/nitrogen (F. Van den Schoor et al., 2009).

Hydrogen flammability limit is wide with Lower Flammability Limit (LFL) of 4% until Upper Flammability Limit (UFL) of 70%. According to Xianshu Lv. (2016), the hydrogen/air explosion accident may happen for a large variation range of hydrogen concentration due to its unique property of wider flammability ranges. If inert can reduce the limit of hydrogen flammability limit, then initial pressure and temperature could increase size or wide the flammability limit from 3% to 76%.

An experiment to know the effect of initial pressure and temperature in hydrogen flammability limit were conducted by Xueling Liu, and Qi Zhang (2014) at various pressure from 0.1 MPa, 0.2 MPa, 0.3 MPa and 0.4 MPa and at various temperature from 21˚C, 40 ˚C, 60 ˚C, 75 ˚C, and 90 ˚C. Base on their calculation on the flammability limit, they found that as initial pressure and temperature were increase, there is decrement of LFL values from 4% v/v to 1.25% v/v. On the other hand, UFL values increase at various initial pressure and temperature steeply.

Table 2.1: Lower flammability limit of hydrogen /air vs initial pressure and temperature recorded from the research (Xueling liu, and Qi Zhang, 2014).

Table 1 – Lower flammability limits of hydrogen–air vs initial pressure and temperature.

Initial pressure (MPa)	21 °C		40 °C		60 °C		75 °C		90 °C	
	LFL(H_2) (V/V)%	Air (V/V)%	LFL(H_2) (V/V)%	Air (V/V)%	LFL(H_2) (V/V)%	Air (V/V)%	LFL(H_2) (V/V)%	Air (V/V)%	LFL(H_2) (V/V)%	Air (V/V)%
0.1	4	96	4	96	4	96	4	96	4	96
0.2	2	98	1.5	98.5	1.5	98.5	1.5	98.5	1.5	98.5
0.3	1.67	98.33	1.33	98.67	1.33	98.67	1.33	98.67	1.33	98.67
0.4	1.25	98.75	1.25	98.75	1.25	98.75	1.25	98.75	1.25	98.75

F. Cammarota et al. (2009) found that when the initial pressure increase, the maximum pressure, P_{max} and deflagration index, K_G increases. The pressure and temperature gives the mixture of hydrogen-air to be more homogenous and increase their diffusivity for the explosion. Thus, it enhances the explosion and this explained why the Lower Flammability Limit (LFL) and Upper Flammability Limit (UFL) of hydrogen are wide as the initial pressure and temperature increases.

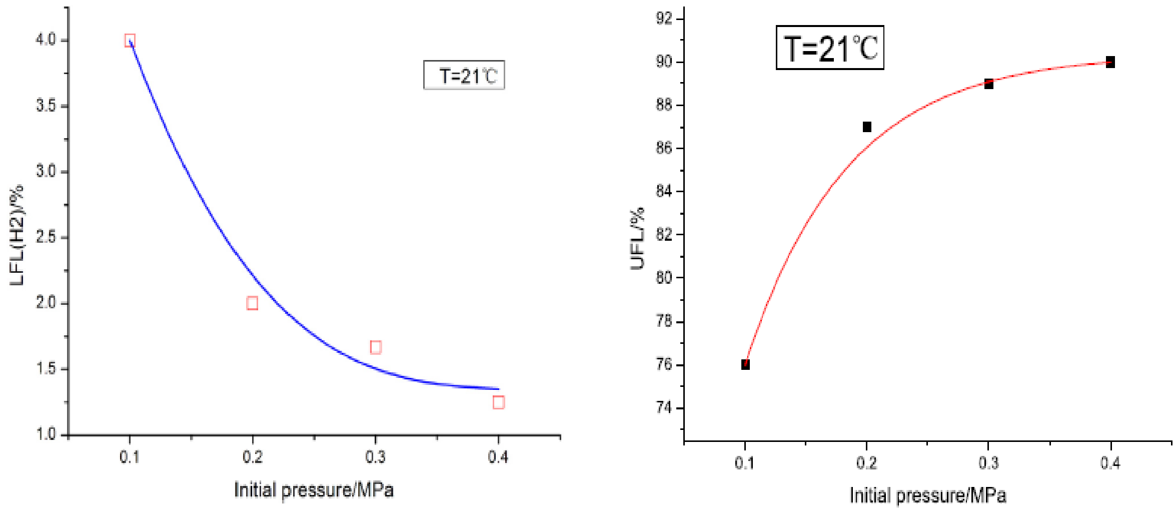

Figure 2.3: Obtained LFL and UFL at various initial pressures and temperature at 21˚C from the research (Xueling Liu, and Qi Zhang, 2014).

From the data in the graph, at initial pressure of 0.4 MPa, the minimum Lower Flammability Limit (LFL) is 1.25% and highest Upper Flammability Limit (UFL) is at 90%. Thus, the severity of hydrogen gas explosion is critical at high initial pressure and temperature.

2.3.2 Fuel concentration

Fuel explosion can be caused by fuel concentration from lean side to rich side. As the concentration increase, the overpressure will increase until it reaches at stoichiometric concentration of 1.0. After the stoichiometric concentration, the overpressure will reduce because of the unbalance of fuel-air mixture content.

Xianshu Lv et al (2016) explained that this may be attributed to the enhancement of combustion reaction caused by the increase of hydrogen concentration, which in turn increased the net rate of volume production of combustion that offset the volumetric flow rate. This can be explained by the lower chemical reaction rate due to the decrease of the equivalence ratio.

They found that the peak overpressure increases firstly and slightly decreases and the extent of reaction has no significant reduction with increasing equivalence ratio once the equivalence ratio is beyond 1.0.

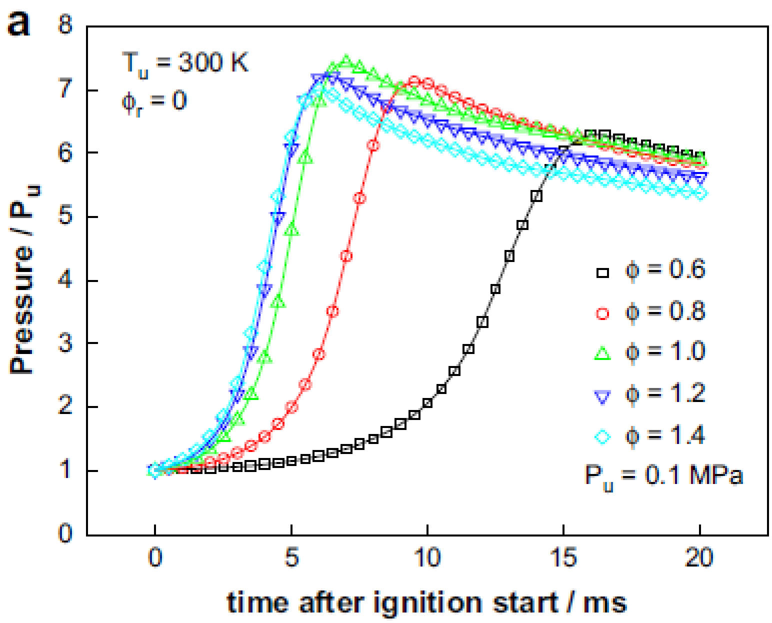

Figure 2.4: Explosion pressure vs time after ignition with various equivalence ratios from 0.6 to 1.4 (Xianshu Lv et al., 2016).

Besides increase in overpressure, increases in fuel concentration will also enhance the flame speed and less time needed for the explosion. Zhu et al. (2010) study the variation in gas concentration in cylindrical vessel. They found that at 10% methane gas concentration, the flame speed increase compare to 8% and 6% methane concentration. This is because the decrease in fuel concentration resulted in a decreased in overpressure and heat released by the reaction which is important for the speed up for the flame.

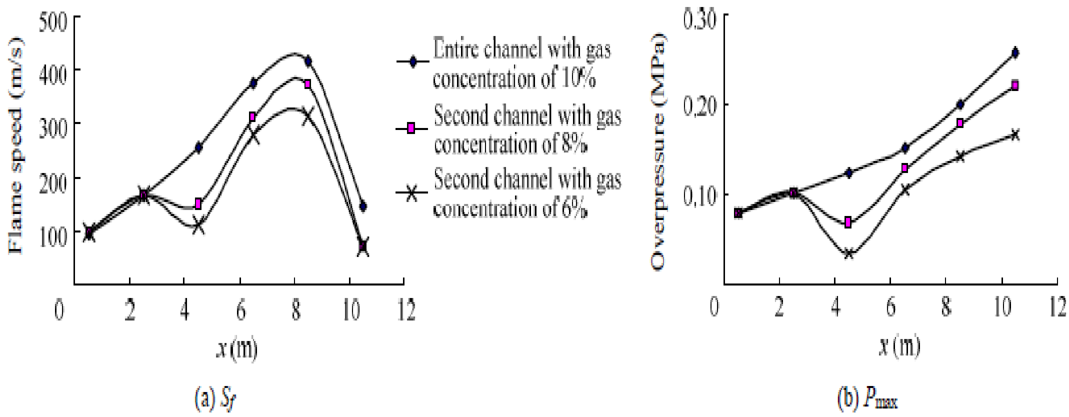

Figure 2.5: Flame speed, S_f , P_{max} vs distance, x travelled by the explosion in cylinder vessel (Zhu et al., 2010).

From the figure, the flame speed and overpressure increase with increase in fuel gas concentration along the cylinder vessel until reach peak of flame speed and overpressure at certain distance. This is because the reflection of a blast wave at the end vessel and also the amount of unburned gas is reduced and thus, the heat released reduced.

2.4 Gas explosion parameters

Several parameters of explosion are:

i. **Maximum explosion pressure, p_{max}:** It depends mainly on the amount of heat generated during combustion (V. Giurcan et al, 2015).

ii. **Flammability limits:** It was defined as a pressure increase of 7% or more than the initial pressure in the vessel (H. J. Liaw et al, 2009).

Lower Flammability Limit (LFL) and Upper Flammability Limit (UFL) were calculated by them using the equation below:

$$L = \frac{-\int_{T_0}^{T} P_L dT + R(T_0 - T) + \alpha e \underline{A}_S \sigma\left(T^4 - T_0^4\right)\Delta t}{\int_{298}^{T} Q_L dT + \int_{T_0}^{298} Cp_f dT - \int_{T_0}^{T}\left[Cp_I - (Cp_I - P_L)\frac{1}{x}\right]dT + \Delta h_c^o - P_{PL}RT} \quad (2.1)$$

$$U = \frac{-\int_{298}^{T} P_U dT + \int_{0}^{298} P_L dT + 0.21 f_U (\Delta h_c) U + RT_0 - (0.79 + 0.21 P_{PU})RT + \alpha e \underline{A}_S \sigma\left(T^4 - T_0^4\right)\Delta t}{\int_{T_0}^{T}(Cp_f - Cp_I)dT + \int_{298}^{T}(Cp_I - P_U)\frac{1}{x}dT + \int_{T_0}^{298}(Cp_I - P_L)\frac{1}{x}dT - 0.21 f_U \frac{1}{x}(\Delta h_c)U} \quad (2.2)$$

The equations above are for use in constant volume system.

iii. **Maximum rate of pressure rise, $(dp/dt)_{max}$:** It depend mostly on the on the rate of heat liberation, influenced by the overall reaction rate in the flame front (V. Giurcan et al, 2015). On the other hand, C. Movileanu et al (2013) stated that it depend on the total initial pressure of the flammable mixture.

The rate of pressure rise during explosion in a closed vessel was correlated to the normal burning velocity by the equation:

$$\frac{dp}{dt} = \frac{3 S_u \rho_u}{R \rho_0}(p_e - p_0) \quad (2.3)$$

where R is the vessel's radius, p_0 and ρ_0 are the initial pressure and initial density of unburned gas, p_e is the end explosion pressure, p and ρ_u are the pressure and density of unburned gas density at time t, γ is the adiabatic coefficient of unburned gas and S_u is the laminar burning velocity.

The peak rate of pressure rise is further used for calculating the deflagration index, K_G of gaseous explosions in enclosures according to the cubic-root law:

$$K_G = (\frac{dp}{dt})_{max} \sqrt[3]{V} \tag{2.4}$$

where K_G was defined by analogy to the deflagration index of dust-air explosion, K_{st} .

2.5 Flame propagation in spherical flame

In this subtopic, more discussion on flame propagation such as Peclet number, Pe, Markstein number, Ma, influence of hydrodynamics instabilities and diffusional-thermal instabilities on flame propagation will be focus to know the characteristics of spherical flame behaviour during explosion.

W. K. Kim et al. (2015) studied the self-similar propagation of expanding spherical flames in large scale gas explosion, have deep discussion on the spherical flame characteristics. They using hydrogen-air, propane-air, and methane-air mixtures as one of their fuel in their study to compare the result obtained from each mixtures.

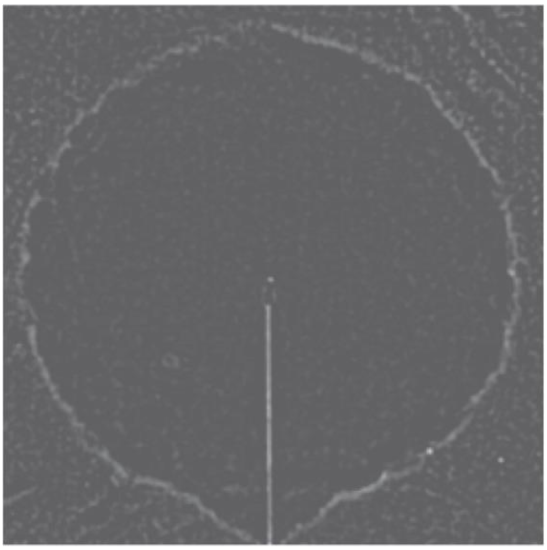

Figure 2.6: Flame front structure captured using MATLAB for hydrogen-air mixture of φ = 1.0 (W. K. Kim et al., 2015).

Critical Peclet number, P_{ec} is defined as the flame radius, r relative to the flame thickness, δ as in the equation below:

$$Pec = \frac{r}{\delta} \qquad (2.5)$$

Where the flame thickness, δ can be measure from the equation below:

$$\delta = \frac{T_{ad} - T_u}{\left(\frac{dT}{dx_{max}}\right)} \qquad or \qquad \delta = \frac{\lambda}{\rho c_p S_L} \qquad (2.6)$$

Peclet number, P_{ec} is used on Markstein number, Ma to evaluate the dependence of both parameters to know the relationship between the critical flame radius and the intensity of diffusional-thermal instability. Markstein number is calculated by equation below:

$$Ma = \frac{L}{\delta} \qquad (2.7)$$

Where L is the Markstein length and in their experiment, Markstein length is computed from the mass burning velocity at the location where the velocity is the maximal.

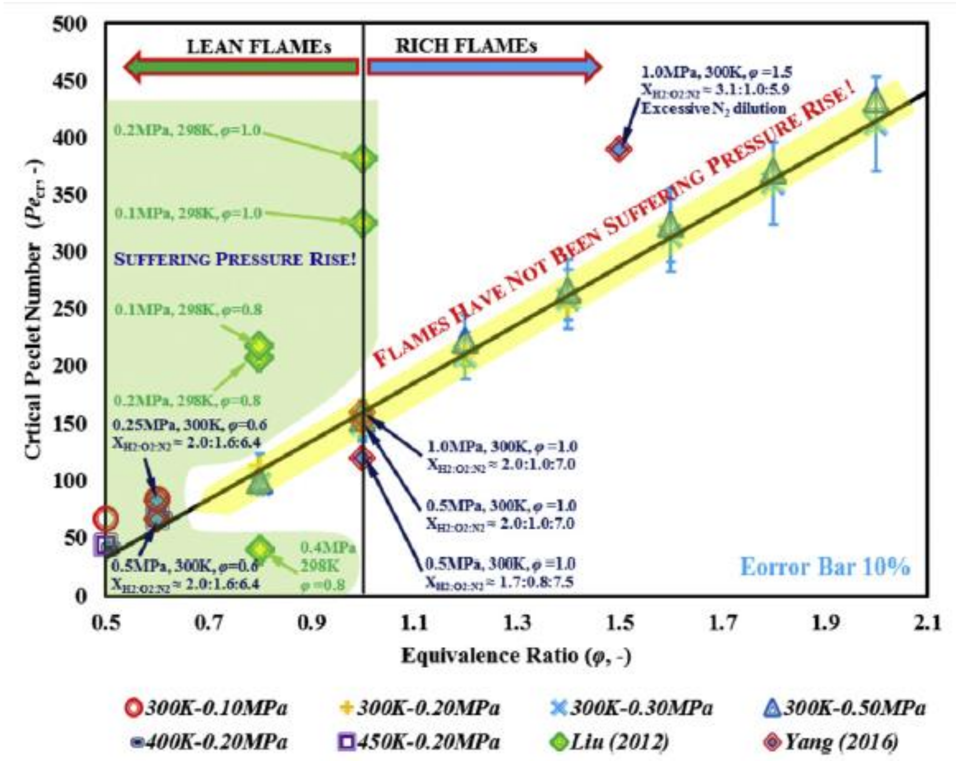

Figure 2.7: Density ratio as the function of equivalence ratio under different initial thermodynamic ambient conditions (Z. Y.Sun et al., 2016).

Z. Y. Sun et al. (2016) explained that at the early propagation of spherical flame, most flames prefer to loss their initial smoothness as they further propagate and then turn to create cellular structure on flame front. The reasons to the behaviour of cellular structure are the hydrodynamic instabilities, and thermal-diffusive instabilities.

Hydrodynamic instability is defined as different density ratio of fuel-air mixture and this lead to density jump across flame front, σ. The larger the density ratio is, the bigger the density jump across flame front, σ and also giving higher intensity of hydrodynamic instability. Kwon et al., (2001) stated that flame thickness, δ is also one factor affecting the hydrodynamic instability.

Thermal-diffusive instability occurred when flame front become non-equilibrium between heat conduction and mass diffusion and this intensity was indicated (Z. Y. Sun et al., 2016) by Lewis number, Le as

$$Le = \frac{D_T}{D_{im}} \tag{2.7}$$

Where D_T is thermal diffusivity of combustible mixture, and D_{im} is the mass diffusivity of limiting reactant. Lewis number, Le is one indicator to reflecting the relationship of thermal diffusive and mass diffusive across flame front.

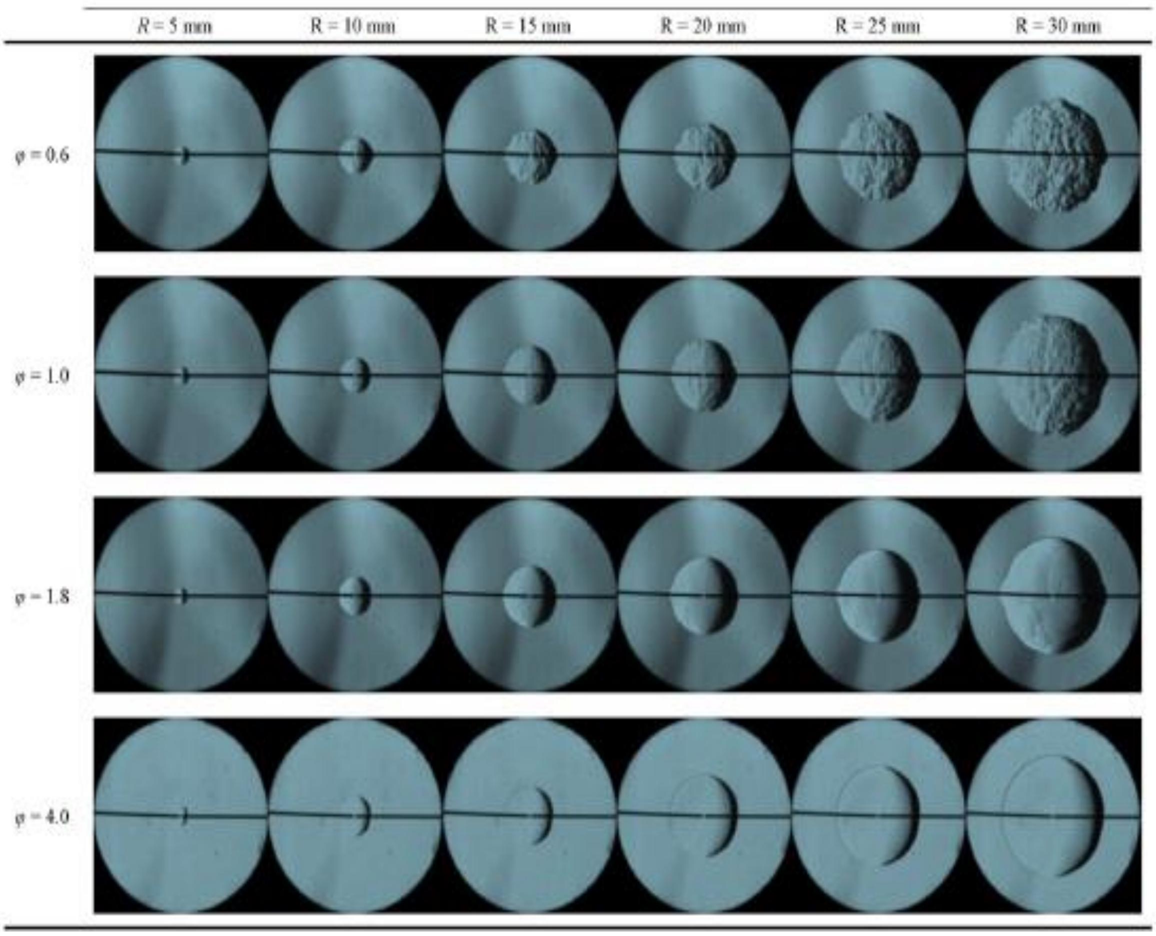

Figure 2.8: Revolution of laminar spherical flames within homogenous hydrogen-air mixtures (Z. Y.Sun et al., 2016).

The figure shows revolution of laminar spherical hydrogen-air flames from lean equivalence ratio, φ = 0.6 to rich φ = 4.0. It clearly shoes that as the propagation radius wider the flame front structure change from smooth to cellular structure.

CHAPTER 3

METHODOLOGY

3.1 Background

This chapter discusses the experimental procedure and the test involved during the explosion test. The experiment will be carry out in a spherical 20 L vessel available in Combustion Lab, Faculty of Chemical and Natural Resources Engineering, Universiti Malaysia Pahang. The spherical bomb diagram, schematic spherical bomb diagram, and overall work flows are shown in Figure 3.1, and Figure 3.2. The experiments start with the explosion of hydrogen-air mixture in spherical vessel test to observe the explosion formation.

3.2 Materials

Pure hydrogen (99.9%) and pure nitrogen (99.9%)

3.3 Equipment

In this study, the experiment was carried out in a standard 20-L explosion spherical vessel according to the ISO6184-1 see Figure 3.1. It consists of an explosion chamber, an electric ignition system, a control unit, a data acquisition system, a release valve, a vacuum pump, an air pump, a view point, pressure sensor, and water inlet and water outlet port. A high voltage electric spark was use to supply ignition energy. The igniter was mounted at the centre of the spherical bomb and a spark energy of 10kJ was delivered by an electric ignition system.

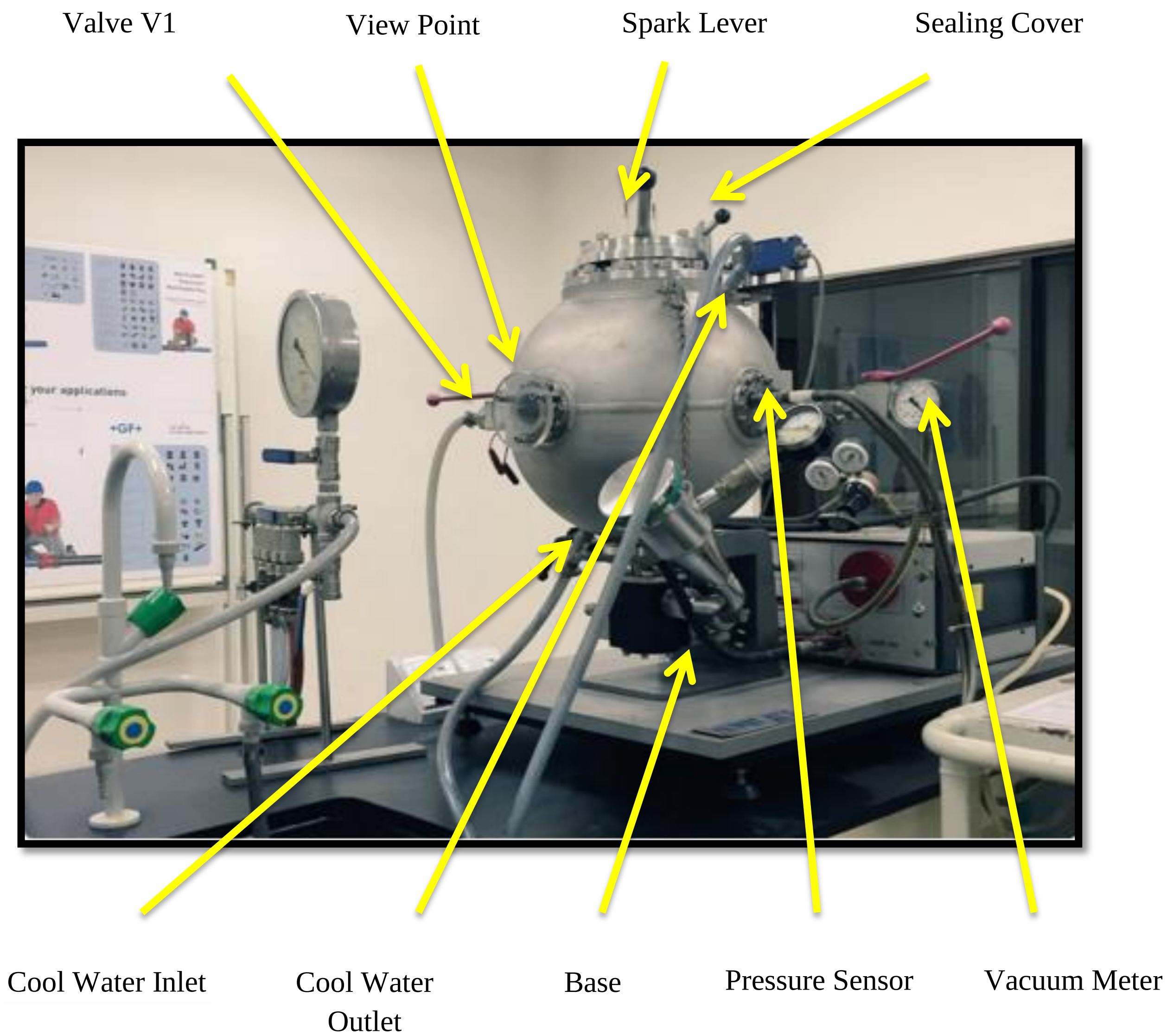

Figure 3.1 The 20-L explosion spherical bomb.

3.4 Method Flow Chart

The simplified methodology is described as in the figure 3.2 below. The experiment will start with the first objective which is to evaluate the effect of equivalence ratio of hydrogen-air mixture in explosion pressure and rate of pressure rise. After conduct the first objective, this experiment will continue to the second objective which is effect of vessel shapes on the explosion pressure and rate of pressure rise.

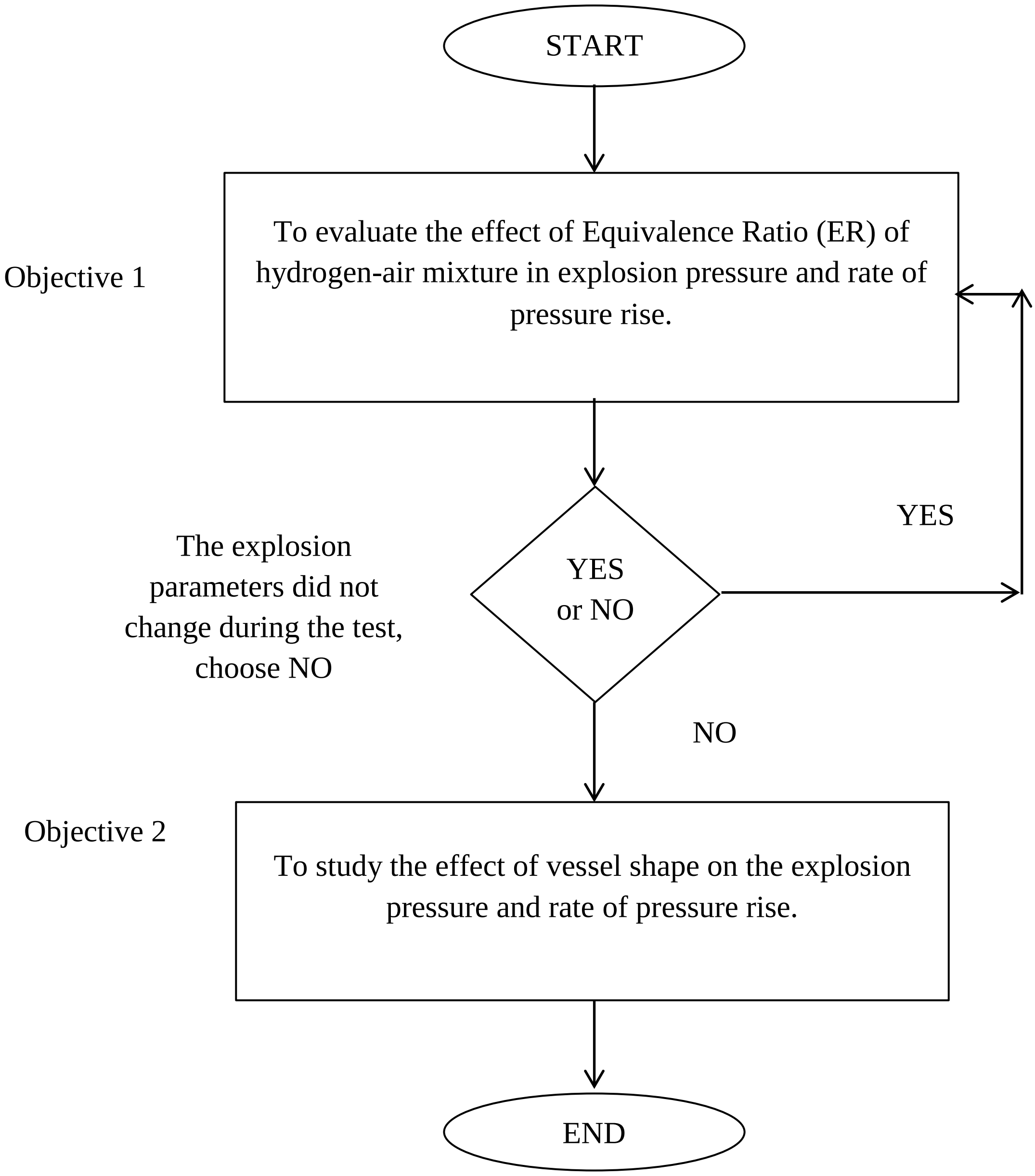

Figure 3.2 Overall work flow chart

3.5 Gas handling and mixing preparation

The range of hydrogen gas concentration will be varied at 12%, 18%, 24% and 30% respectively. The partial pressure method of mixture preparation will be applied to add the flammable gas to a vacuum and then adds air to approximately at 1 bar. The explosion will be carried out after a delay of about 10 min for the mixture to be homogenous. This method of mixture preparation ensures complete mixing, as the initial vacuum condition rapidly disperses the fuel added and subsequent addition of air takes place under still very low pressure; together with the turbulence from the air injection, this ensures rapid mixing. The mixture composition will be controlled to an accuracy of 10 Pa (0.01% of composition). To check the homogeneity of the hydrogen/air mixtures, gas analysis using gas chromatography will be carried out. As part of the experimental programme, three repeat test will be performed at each condition to demonstrate a good reproducibility, with peak pressures varying by less than ±5% in magnitude.

Table 3.1: Composition of hydrogen used in experiment

Fuel	**Hydrogen, H_2**
Equivalence Ratio, Ø	**Vol/vol %**
0.4	12
0.6	18
0.8	24
1.0	30

The partial pressure methods used to calculate pressure of hydrogen and air in experimental work shown in Equations (3.1) and (3.2). The pressure calculated are listed in Table 3.2.

$$1000\ mbar\ x\ hydrogen\ concentration\ \text{(Vol/vol \%)} = \text{Pressure}_{Fuel} \quad (3.1)$$

$$1000\ mbar - \text{Pressure}_{Fuel} = \text{Pressure}_{Air} \quad (3.2)$$

Table 3.2: Calculated pressure for hydrogen and air for mixing preparation

Equivalence Ratio, Ø	Concentration (Vol/vol %)	Hydrogen pressure, mbar	Air pressure, mbar
0.4	12	120	880
0.6	18	180	820
0.8	24	240	760
1.0	30	300	700

3.6 Methodology

3.6.1 Initial preparation

i. Purge the vessel with nitrogen gas to remove the impurities gas in the vessel.
ii. Connect the spherical bomb to the data logger (KSEP system) at the personnel computer.

3.6.2 Mixing preparation

i. Evacuate the vessel to 0 bar using external vacuum pump.
ii. Transfer the fuel to the spherical bomb via gas transfer tube.
iii. Disconnect the fuel hose. Open valve, V1 to allow the ambient air filling the vessel until reach the atmospheric pressure.
iv. Closed valve 1, V1 to let the hydrogen gas and air to be premixed in 10 minutes.
v. Collect sample after 10 minutes premixed at 15µml at valve 1, V1 for gas chromatography (GC) analysis to test the concentration and homogeneity.

3.6.3 Ignition & explosion test

i. Once step 1 & 2 completed, push the start button at the ignite system and record the data at the recording unit.
ii. After the logging is completed, safe the raw data for further analysis.

3.6.4 Post test

i. Open valve 1, V1 slowly to release pressure until reach the atmospheric pressure.
ii. Purge the vessel for 10 minutes to remove residue fuel vessel and combustion product.
iii. Open valve 1, V1 for air circulation using vacuum.
iv. Stop the vacuum pump and disconnect from the test vessel and ready for the next run.

CHAPTER 4

RESULT AND DISCUSSION

4.1 Introduction

This chapter will discuss the result obtained in the experimental work. The elements that will be discussed were explosion parameters obtained which are maximum explosion pressure, P_{max} , maximum rate of pressure rise, $(dP/dt)_{max}$, maximum explosion index, K_{max} , and explosion parameters in different vessel shape.

4.2 Explosion parameters in a spherical vessel

4.2.1 Maximum explosion pressure, P_{max}

a) Maximum explosion pressure, P_{max} at various concentration

Maximum explosion pressure, P_{max} was depends mainly on the amount of heat generated during combustion (V. Giurcan et al, 2015). The amount of heat generated also depends on the amount or concentration of the hydrogen mixed in air. This theory would help in determining the safe amount of hydrogen in the air to avoid massive explosion in industry.

Three experiments were conducted to analyse the explosion pressure at it's equivalence ratio, φ = 0.4, 0.6, 0.8, 1. From the experiment tabled in Figure 4.1, the explosion pressure value at different equivalence ratio is 2.8 bar at 0.4 v/v%, 4.3 bar at 0.6 v/v%, 5.1 bar at 0.8 v/v% and 5.6 bar at 1 v/v%. This phenomenon can be explained by the lower chemical reaction rate due to the decrease of the equivalence ratio (Lv Xianshu et al, 2016). Lower chemical reaction would have low amount of heat generated for the combustion. Thus, at higher equivalence ratio, the maximum explosion pressure would be higher and more severe.

This study only focused on concentration from lean to stoichiometry side. As at low concentration, the amount of hydrogen content in air was low which is at 4 v/v% and this result can show that hydrogen will able to cause an explosion even at low hydrogen content in air.

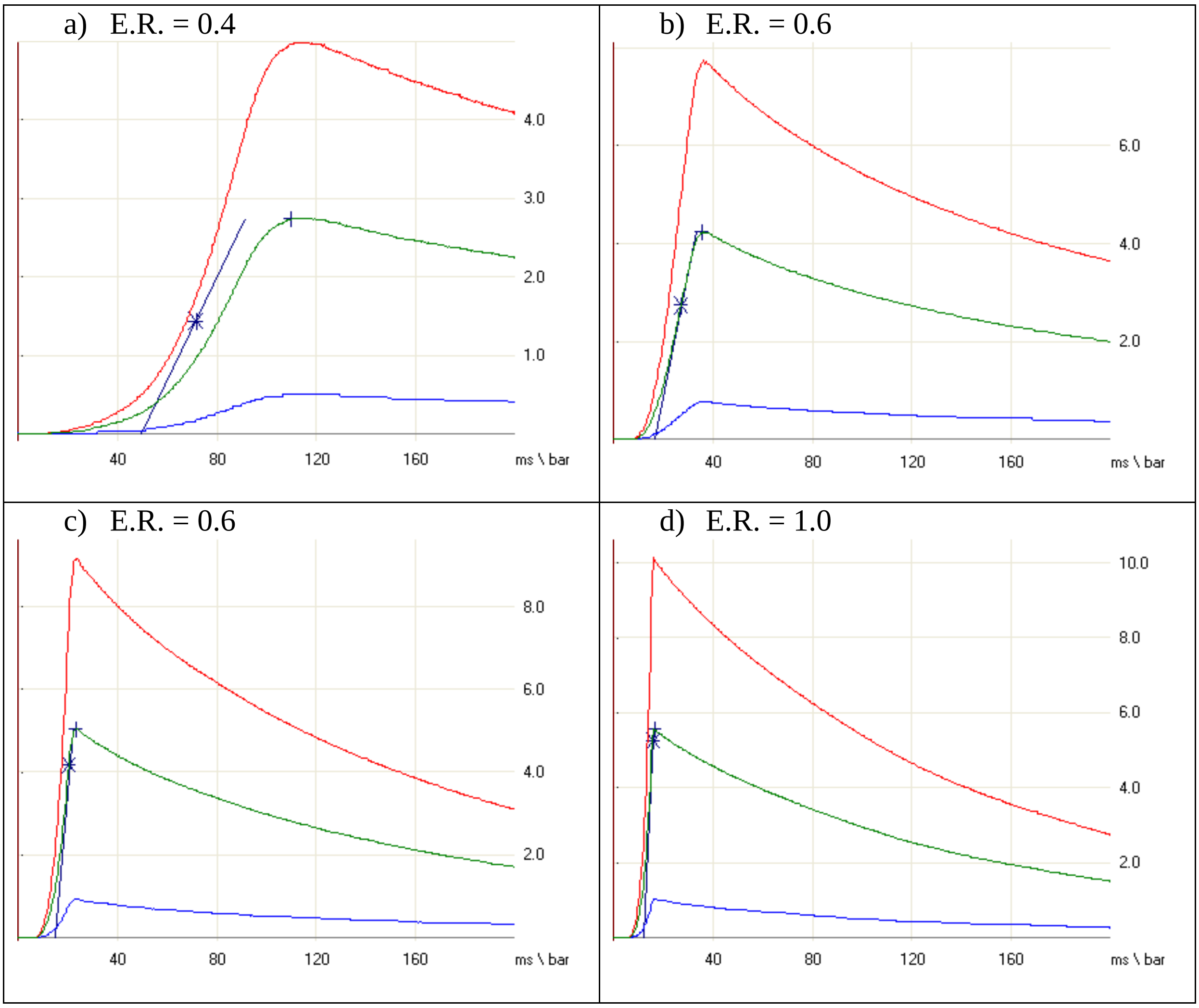

Figure 4.1: Maximum explosion pressure, P_{max} at a) E.R. = 0.4, b) E.R. = 0.6, c) E.R. = 0.8, and d) E.R. = 1.0.

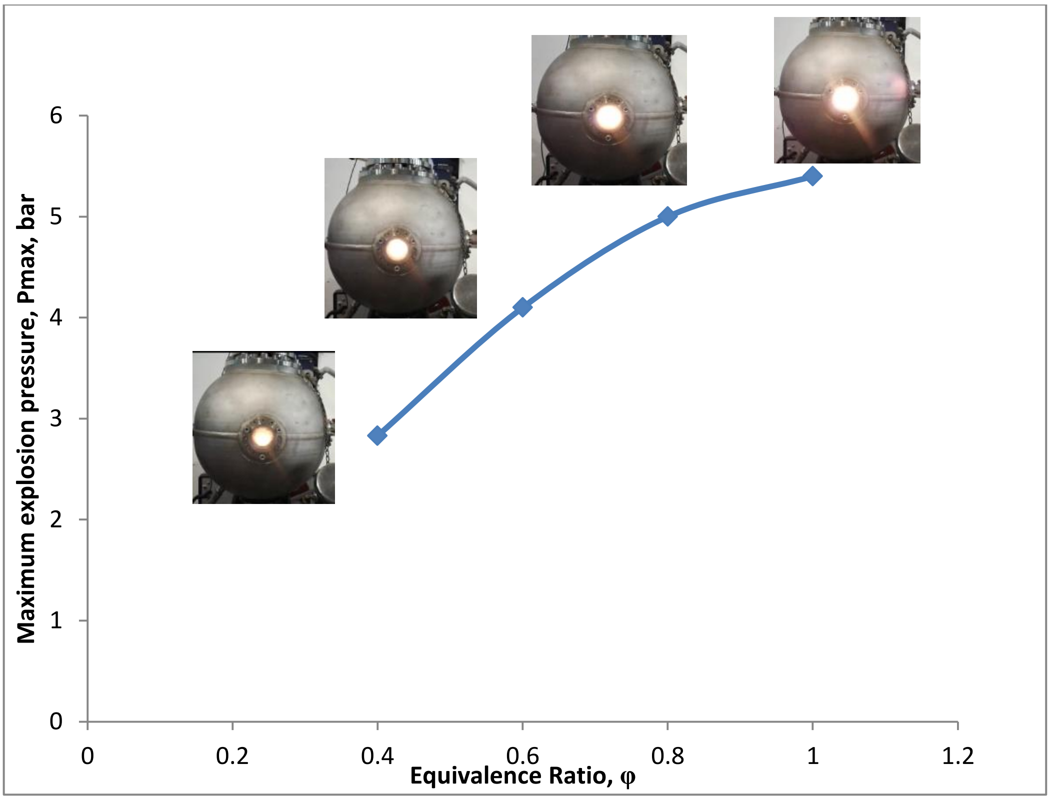

Figure 4.2: Summarized maximum explosion pressure vs equivalence ratio

Figure 4.3 shows the increasing value of explosion pressure vs equivalence ratio and the explosion development in the spherical bomb. P_{max} obtained increases as fire intensity in each equivalence ratio was getting bigger. The increases amount of hydrogen concentration in the mixture give much mass of fuel and this lead to the increases in combustion reaction for the explosion.

During explosion, the total unburned gas become less as the mass burning rate increase at each equivalence ratio, φ and enhance the flame radius range in the spherical flame. As the flame front propagated away from the ignition center, the flame structure evolved from laminar to turbulent (cellular) structure. From this experiment, the light intensity showed in the figure 4.3 describe the flame propagation occurred from laminar at lean to turbulent at stoichiometric concentration.

These phenomenon resulting to the increases of heat release and the explosion pressure, P_{max} increases as the wide flame front that burning much amount of unburned gas reached the wall. This would explain that the hydrogen-air mixture explosion is more severe at stoichiometry than at low concentration.

b) Maximum explosion pressure, P_{max} with response to time

Maximum explosion pressure, P_{max} has clear correlation with time of ignition. This can be explained by the level of homogeneity. Once the hydrogen mixed with air, the areas of homogenous mixture in constant volume vessel are different. This means that the ignitor sparks would have a probability to not in contact to the complete mixture of hydrogen-air. As the gas mixture circulate within the constant volume, the chance for the spark to contact to the mixture become greater. Thus, less time needed for the ignition.

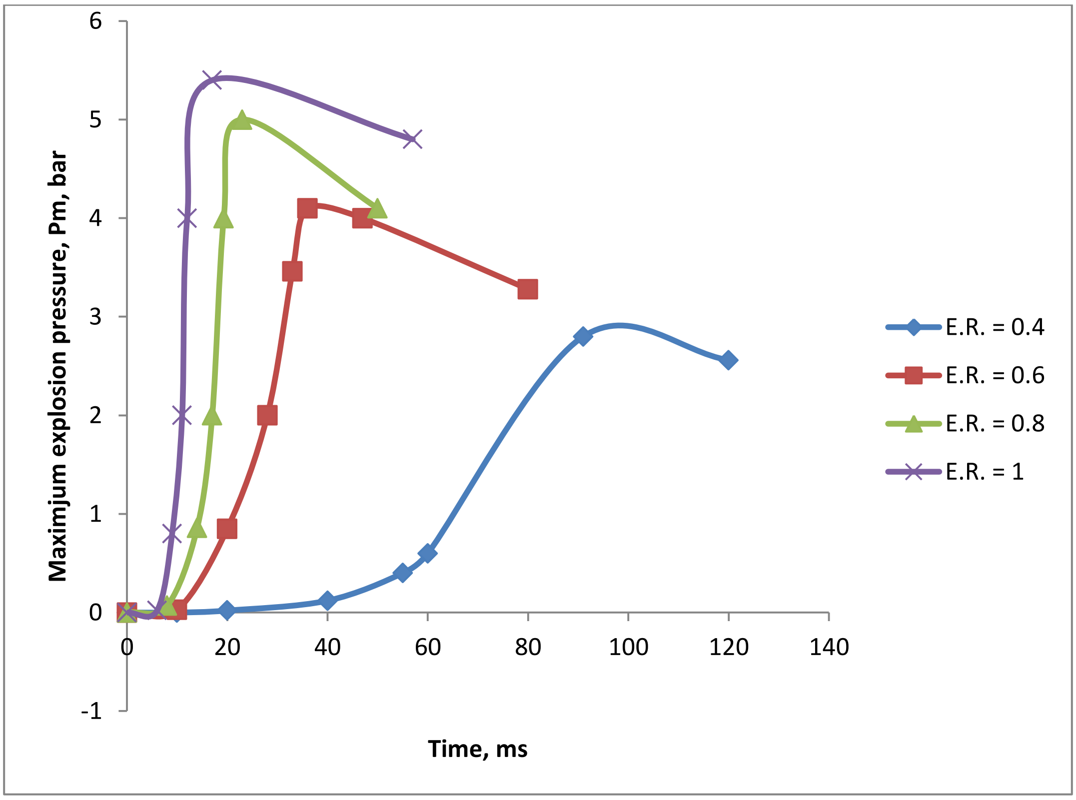

Figure 4.3: Time taken to reach maximum explosion pressure at various equivalence ratios.

Figure 4.4 shows that the maximum explosion pressure of 5.4 bar occurred at 17 ms for stoichiometry hydrogen-air mixture (E.R. = 1) while 2.56 bar at 120 ms for lean concentration or E.R. 0.4. It was expected when increase the hydrogen concentration, the fast reaction discovered. This was due to increase amount of hydrogen (fuel) in the air. For lean hydrogen concentration, the thermal-diffusive instability influences the flame stability of lean hydrogen-air flames rather than stoichiometric flames (Z. Y. Sun et al., 2016). This describe that thermal-diffusive instability was dominantly affected by fuel concentration. Thus for stoichiometric concentration, the diffusivity of hydrogen into the flame propagation in the spherical flame was great.

In an experiment conducted by Xianshu Lv et al (2016) said that the previous studies indicated that the flame front and the overpressure increase significantly with the hydrogen addition. In addition,(A.E. Dahoe, 2003) stated that, the duration of an explosion in a 20-L sphere is long enough to allow the flame ball to rise in the vessel due to the buoyancy.

4.2.2 Rate of pressure rise, dP/dt at various concentrations

Rate of pressure rise has, dP/dt same correlation with maximum explosion pressure, $P_{max.}$ High energy content in high concentration of hydrogen brings chemical reaction to maximum until reach peak rate of pressure rise at 1410.3 bar/s at equivalence ratio, φ of 1. Xiao H. (2013) stated that the unique characteristic of hydrogen itself is due to its high reactivity, and diffusivity which can lead to pre-ignition flashback, and explosions. The upwards trend showing that lean side of hydrogen-air mixture explosion could release high pressure during the explosion and even the concentration of hydrogen (fuel) was less than air concentration in the mixture. Hydrodynamic instability become fiercer in stoichiometric concentration and this enhance the dP/dt as the density ratio across the flame front is increase.

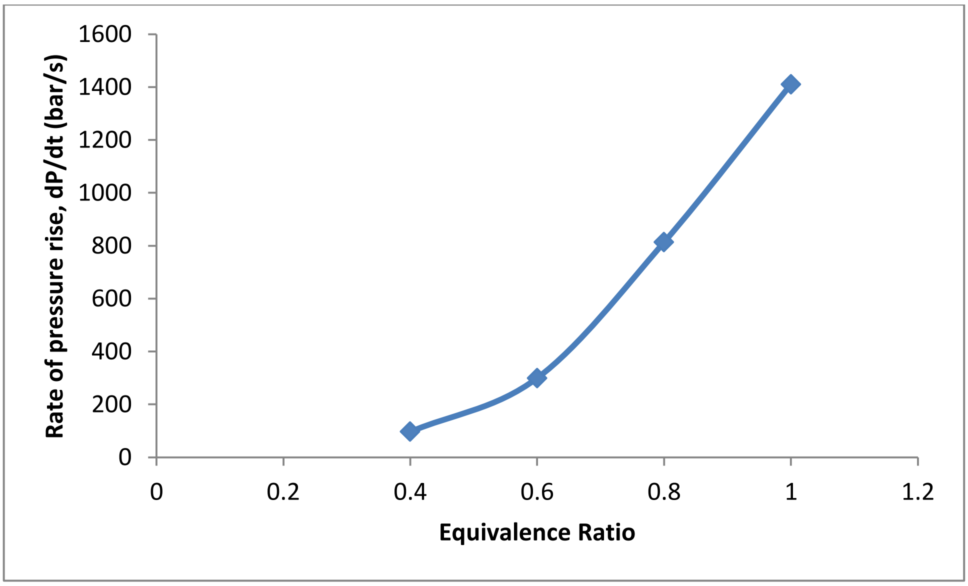

Figure 4.4: Rate of pressure rise at various concentrations.

4.3 Comparison in explosion parameters between spherical shape and cylindrical shape

This subtopic will discuss the comparison between the explosion parameters in different vessel shape between spherical and cylindrical vessel shape in order to know the severe condition when explosion happen in closed vessel.

4.3.1 Maximum explosion pressure, P_{max}

The design specifications of those three vessels are as in the table below.

Table 4.1: Design specification for different vessel shapes

Vessel	**Diameter (m)**	**Length (m)**	**Volume (m^3)**
2-in	0.05	0.1	0.0039
4-in	0.1	5.1	0.04
Spherical shape	0.34	-	0.02

Different diameter sizes were choose to study the flame propagation mechanism and to evaluate the severity of explosion in different diameter size. An experiment conducted by A. E. Dahoe (2005) stated that the reason he choosing a small volume vessel was to achieve a significant amount of pressure build up before buoyancy effects would manifest themselves.

Figure 4.6 shows the maximum explosion pressure in different vessel shapes. Figure 4.6 shows 2-in pipe with low volume have 1.28 times higher maximum explosion pressure, P_{max} at 9.1 bar compare to other explosion pressure value in 4-in pipe and 1.68 times cylindrical vessel. The ascending order for the explosion pressure in those three vessels were $V_{2\text{-in}}$ (9.1 bar) > $V_{4\text{-in}}$ (7.1 bar) > $V_{Spherical}$ (5.4 bar). In small diameter pipe, the flame propagates rapidly to producing more turbulence and giving a rapid in flame acceleration (S. Z. Sulaiman, 2015). Meanwhile, in larger diameter, the heat loss to the wall increases to give less magnitude of turbulence intensity. This was due to the quenching effect in large diameter effect the explosion pressure. Rate of reaction for fuel to react with the flame front is lower in large diameter because flame propagation need to burn the unburned gas at the wall and this cause the release of explosion pressure in the vessel was less.

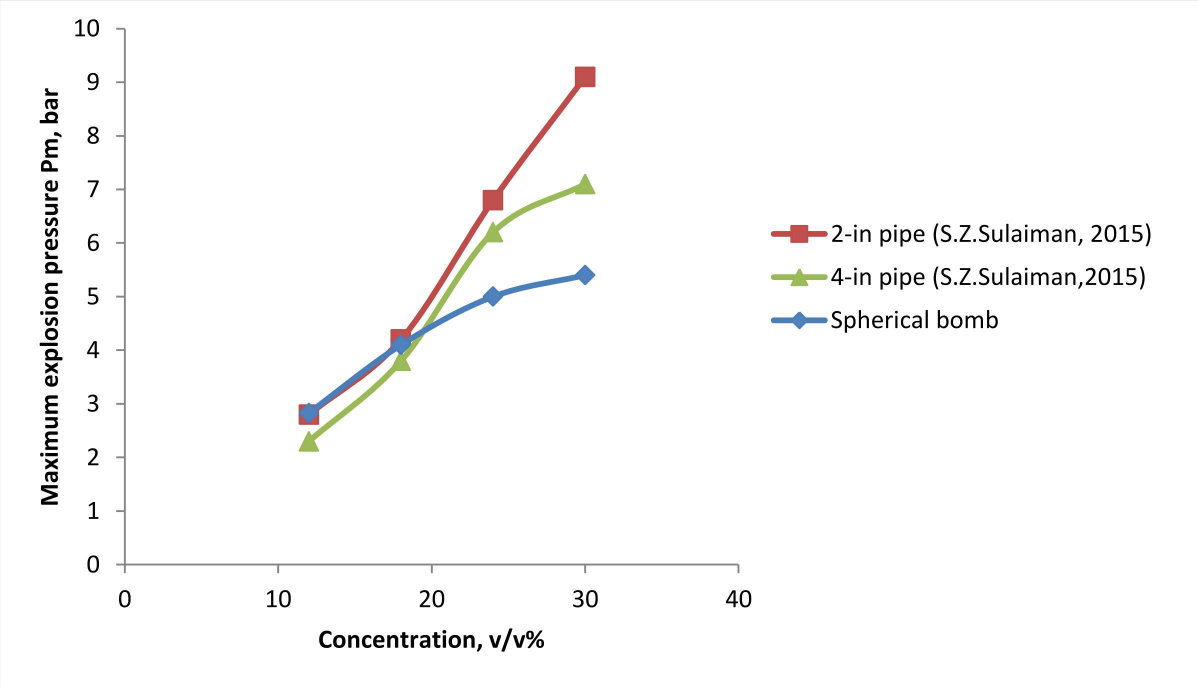

Figure 4.5: Maximum explosion pressure vs concentration at various vessel shapes.

4.3.2 Rate of pressure rise, dP/dt

Figure 4.7 shows the spherical vessel have higher rate of pressure rise, dP/dt at 1410.3 bar/s at stoichiometry 1 compare to 2-in pipe at 204.93 bar/s and 4-in pipe at 124.60 bar/s. High rate of pressure release during the explosion shows that spherical vessel have higher severity compare to 2-in pipe and 4-in pipe. Hydrodynamic instability become fiercer in stoichiometric concentration and this enhance the dP/dt as the density ratio across the flame front was increase. In a spherical flame, the surface burning area was much larger and cause the mass burning rate of hydrogen-air to the flame front were much faster as the diffusivity was fast. Thus, this mechanism influence the flame propagated from laminar to turbulent.

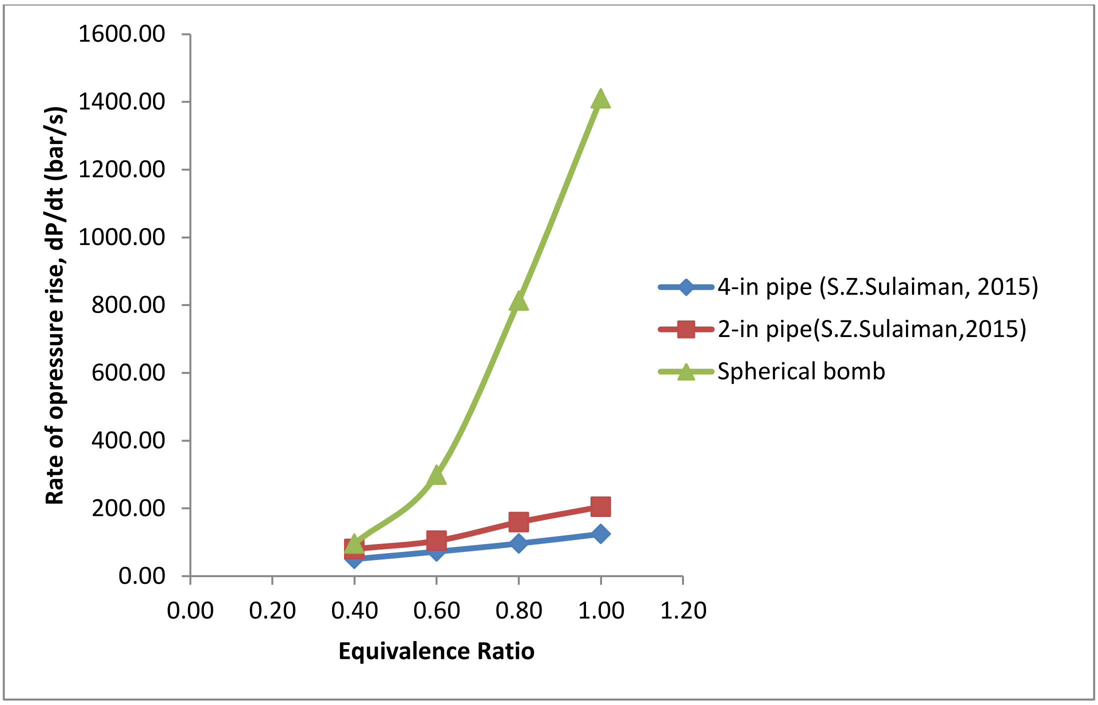

Figure 4.6: Rate of pressure rise vs Equivalence Ratio at various vessel shapes.

4.4 Mixture reactive index, K_G in spherical and cylindrical vessel

The maximum rate of pressure rise, dP/dt and mixture reactive index, K_G is two important explosion characteristics of mixture. These two characteristic would use to quantify the potential severity of an explosion. K_G values were calculated and tabulated in the table 4.2 below. M. Faghigh et al (2016) related the K_G and dP/dt in the equation below:

$$K_G = (\frac{dP}{dt})_{max} \times V^{1/3}$$

V is the volume of the vessel.

Table 4.2: Mixture reactive index, K_G in different vessel

	K_G (bar.m/s)		
v/v %	**Spherical**	**2-in pipe**	**4-in pipe**
12	17.92	12.66	17.29
18	69.76	16.43	24.78
24	190.82	25.09	32.98
30	378.12	32.26	42.61

Overall reading shows that the K_G index increasing at higher concentration in each type of vessel. This was encouraged by the amount of hydrogen was increase. Explosion in spherical vessel has higher K_G index at 378.12bar.m/s for stoichiometry concentration of 30v/v%. 4-in pipe has greater K_G index than 2-in pipe because the bigger volume vessel can hold much hydrogen concentration compare to hydrogen concentration in 2-in pipe. On the other hand, spherical vessel would give bigger value compare to the other cylindrical vessel. The flame propagation in spherical vessel was much greater than 4-in pipe because the flame burned all the gas mixture and produced more rate of pressure rise, dP/dt. Even the volume of spherical vessel was small compare to 4-in pipe, but the flame propagation determines the severity of the explosion in vessel. From the table, explosion in spherical vessel has higher severity followed by 4-in pipe and 2-in pipe.

CHAPTER 5

CONCLUSION AND RECOMMENDATION

5.1 Conclusion

This experiment have been carried to achieve the objectives which are to evaluate the effect of Equivalence Ratio (ER) of hydrogen – air mixture in explosion pressure and rate of pressure rise and to study the effect of vessel shape on the explosion pressure and rate of pressure rise. The conclusions below summarized the finding from this experiment.

i. Maximum explosion pressure, P_{max} of hydrogen-air mixture explosion increase with increase of hydrogen concentration. The highest explosion pressure is at hydrogen stoichiometry concentration in air at 30 v/v% with P_{max} = 5.4 bar.

ii. Rate of pressure rise, dP/dt of hydrogen-air mixture explosion increase with increase equivalence ratio. The amount of heat loss to the wall in spherical vessel shows the severity of the explosion at different concentration of hydrogen in air.

iii. 2-in pipe which is small in 0.05 m in diameter, have higher maximum explosion pressure, P_{max} compare to 4-in pipe with 0.1 m in diameter. Pipe diameters were varied to know the explosion pressure in these pipes.

iv. Comparison of explosion parameters between spherical vessel and cylindrical vessel shows that 2-in pipe with smaller volume give higher maximum explosion pressure while spherical vessel give higher rate of pressure rise which is more severe than the others two cylindrical vessel.

5.2 Recommendation

Recommendations for improvement for future experiment are as below:

i. The vacuum pressure line connected from main box to the spherical bomb need to be clean and replace with new and clean cotton to increase efficiency of carbon metal absorption.

ii. Operate the fuel/air inlet valve fully to avoid sensitivity of pressure in the system.

iii. A digital flow meter at inlet fuel/air line need to replace the analog flow meter. This is to avoid an error of fuel/air inlet concentration as digital flow meter can help give accurate amount of concentration to the mixture.

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