Effect of inhibitor gases on hydrogen flame propagation in a confined tee pipe (Part I)

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A B S T R A C T

Hydrogen is a promising fuel for the future. In recent years it has been successfully utilised in industries, particularly in refineries and petrochemicals. In previous studies, the effect of inhibitors on hydrogen explosion behaviour has been investigated in different systems, yet only scarce data are available. Therefore, experimental study is carried out to investigate the effects of argon, nitrogen and carbon dioxide on hydrogen/air explosion in a branched pipe configuration. The fuel/air mixtures were ignited at three different ignition positions, A, B and D. The results show that, when ignited at the furthest distance (position A), the tee junction area is most vulnerable to the critical pressure impact of gas explosion. However, no similar trend was observed at the other ignition positions. In addition, mixtures with the compositions 95% H2–2.5% Ar–2.5% N2/air, 95% H2–5% N2/air, H2/air and 95% H2–5% Ar/air showed higher risks due to the higher diffusivity ratio and the associated rate of pressure rise. This phenomenon is highlighted in the discussion part of this paper. The results show that mixtures with CO2 lead to lower severity than other compositions (~50% reduction), as the average recorded maximum flame speed for this particular mixture was lower at all of the ignition points. This suggests that the effectiveness of the inhibitors should be in the order of Ar < N2 < CO2.

1. Introduction

Hydrogen has been promoted for several decades as an energy carrier of the future. As a secondary energy carrier with a lower heating value of 120 MJ/kg and density of 0.0899 kg/m³, it can be used in many applications, particularly in refineries and petrochemicals [1]. However, hydrogen explosion hazards have constantly been a concern for its storage and transportation through pipelines; it is essential to address safety issues related to pipeline gas carriers by characterising and adequately quantifying their explosion behaviour to protect against the propagation of unwanted combustion phenomena such as deflagrations and detonations (including decomposition flames) [2–4]. Previous research has reported that hydrogen is susceptible to deflagration-to-detonation transition (DDT) [5]; it is important that this parameter can be calculated and predicted with a reasonable degree of accuracy towards the end goal, which is control over flame acceleration and the process of DDT. An extensive amount of work has therefore been devoted to understand the phenomena related to flame acceleration and DDT in pipes. These previous works have discussed and illustrated the fact that the changes of the flame-front shape and propagation in a tube, depend on numerous parameters, such as aspect ratio, initial pressure, open or closed tube ends, and equivalence ratio of the mixture [6]. The dynamics of the flame mechanism during the development of premixed hydrogen–air flames at various equivalence ratios undergoes a complex flame inversion and exhibits more distinct characteristics than with other gaseous fuels [7]. Rai et al. [8], in their report on the high-pressure release of hydrogen gas inside a 6-m-long horizontal tube, indicated that the pressure starts to build up as the flame propagates closer to the jet velocity, and that deflagration that takes place closer to the jet velocity results in higher pressure peaks [8]. Petukhov et al. [9] furthermore examined shock-wave explosion propagation of hydrogen–oxygen mixtures. They observed that the instability of flame propagation in a non-stationary combustion front results in the formation of disturbances, waves and streams prior to the flame front. Subsequently, flame intensification creates secondary combustion or explosions,

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exceeding the value of the Chapman–Jouguet conditions for stationary detonation [9]. Numerical simulation and experimental studies on flame propagation in a duct with a 90° curved section also showed a good agreement on basic flame-propagation physical phenomena such as the tulip-shaped flame, flame shedding, pressure evolution trends, flame-propagation speed profiles and vortex development at the bend [4,10]. However, hazard assessments of these phenomena are still debatable, even though these concerns have long attracted researchers’ attention. It is essential that reliable and cost-effective prevention and mitigation methods are available to practitioners and researchers to assess hydrogen explosion hazards in the pipeline in order to ensure that no loss of containment or hydrogen leakage occurs during transportation. One possible means for mitigating the hazards associated with unintended hydrogen release or DDT in the pipe is the use of inhibitors [11].

Previous investigations have been more focused on the effect of the presence of inhibitors in hydrocarbon and hydrogen fuels on the laminar flame propagation velocity, laminar combustion velocity, Markstein length, flame stability and maximum combustion pressure [12,13]. These investigations have analysed different combustion properties, but there are many unresolved issues regarding the quantitative relationship between the experimental scales and real practices. These are due to various assumptions regarding different concentrations of fuel–inhibitors, tube/vessel system applications and condition boundaries in terms of the response to the inhibitors’ effects. For example, Qiao et al. [14] studied the flame-stretching effects on the laminar burning velocities of H2–O2-diluent (N2, Ar or He) in a small-scale system (a spherical windowed chamber). They reported that these flames were very sensitive to flame stretching, exhibiting a ratio of unstretched-to-stretched laminar burning velocities in the range of 0.6–3.0, with corresponding Markstein numbers in the range ±3 to 7. They also indicated that diluents, i.e., N2, Ar, and He, tend to have a significant effect on the preferential-diffusion/stretch interactions due to their dynamic and kinetic characteristics, i.e., argon and helium have higher laminar-burning-velocity ratios compared to nitrogen, based on their higher flame temperatures and transport rates [14]. A further study on the flame propagation of H2–O2–N2 using an International Thermonuclear Experimental Reactor (ITER) showed that both overpressure and laminar flame speed increase as the initial pressure and temperature increased. It was also indicated that this condition was most preferable for a higher nitrogen concentration [15]. On the other hand, Di Benedetto et al. [16] made a different observation on the effectiveness of CO2 in H2–O2–N2–CO2 mixtures. They suggested that the burning velocity of the fuel–inhibitors mixtures is mainly due to their kinetic mechanism, as the rapid reaction of O, H, CO, HCO, OH, HO2 and CO2 has a negligible effect on the transport rate. However, the explosion behaviour of stoichiometric H2–O2–N2–CO2 mixtures in NTP (normal temperature and pressure) condition showed significant stability in the flame; this could suggest that there would be no ignition if CO2 presence were higher than 40%. The explosion was suggested to be mainly affected by CO2 and O2 concentration [16]. Moreover, CO2 has a significant radiation and dilution effect on quenching and blow-off limits, as reported by Shih [17]. It is worth noting that the effectiveness of all of these inhibitor gases on Markstein numbers has been insignificant in these studies, due to highly similar dynamic properties [13,14,16]. Despite such extensive studies, the discussion on the influence of the inhibitors concerning explosion in the pipe is still evolving, particularly in relation to the unsteady propagation of flames under branched-pipe conditions.

The point at which ignition takes place is an important factor during the initial stages of combustion [18]. When the ignition source is further from a sealing flange or vessel wall, the flame will have a longer period to spherically propagate, leading to initially higher overall flame speed and rate of pressure rise. Changes in these initial conditions will also affect the continuation of combustion further along the pipe, particularly when the pipe has different configurations, i.e., obstructed, bending and branched. Investigations of fuel transfer in pipes have shown that 36.5% v/v hydrogen in air has the potential to develop DDT [19,20]. Blanchard et al. [21] have also reported that the ignition position has a significant effect on DDT development in hydrogen/air explosion. In their study, the shortest run-up distance to DDT relative to the end pipe was recorded when the ignition source was placed 0.7 m from the pipe ends. From the discussion above, it can be postulated that there is a direct relationship between the changes of ignition positions and the presence of inhibitors on the pressure-development mechanism and flame dynamics during gas explosion inside the branched pipe – this is the focus of the current research.

The present paper reports the explosion pressures at different ignition positions, the maximum rate of pressure rise and severity magnitude of explosion of hydrogen–air mixtures. The effect of inhibitors in this context is also explored; this is achieved by the close examination of pressure waves at various points in the branched pipe configuration.

2. Methodology

In this study, a tee-pipe rig configuration was used. The pipe length was 4.32 m with a diameter of 0.1 m. The junction pipe segment was 1.375 m long (refer to Fig. 1). The pipe was made up of a number of segments, ranging from 0.5 to 1 m in length, bolted together with a gasket seal in between the connections and blind flanges at both ends.

Measurements of flame speed were made using an array of exposed-junction, mineral-insulated, type-K thermocouples CHAL OMEGA (accuracy ±0.001 s) along the centre-line of the entire pipe (indicated by T1–T8 in Fig. 1). The flame-speed data were generated from the thermocouple flame arrival times, the time of travel between two adjacent thermocouples, and the distance between them. The flame speeds were plotted at the position midway between the thermocouples, or in the case of the first flame speed at the time between the spark and arrival at the first thermocouple. This technique does not measure the flame temperature as the thermocouple junction was too large (~0.5 mm); however, there is no dead time and the flame is detected as a sudden increase in temperature from a near-ambient base temperature. The thermocouple flame arrival time in the pipe was taken to be the first point at which the reading started to rise. For the thermocouples in the pipe, the arrival time was delayed by a pre-compression wave ahead of the flame (and the associated high flow velocity around the thermocouple), which gave rise to two distinct gradients on the thermocouple trace. In this case, the point at which the second (steeper) gradient became apparent was taken as the flame arrival time. Pressure measurements were taken using seven piezoresistive pressure transducers (P1–P7 in Fig. 1) to continually measure the pressure development and rates of pressure rise inside the test pipe. The pressure transducers were Keller Series 11, with accuracy of ±0.001 s. The data generated were collected using a 34-channel transient data recorder manufactured by NI CompactDAQ. Information about the accuracy and reproducibility of detected parameters can be found in Wang et al. [22]. The ignition positions and each location of the sensors are presented in Table 1.

The mixtures of H2–Ar/air, H2–CO2/air, H2–N2/air, H2–Ar–CO2/air, H2–Ar–N2/air and H2–N2–CO2/air with a constant ratio (95:5 = H2: inhibitors) were applied at a stoichiometric equivalent ratio (φ = 1) by considering hydrogen as the primary gas. The partial pressure method of mixture preparation adds the flammable gas to a vacuum (VACUUBRAND RE 2.5) and then adds air to
approx. 101,325 Pa. The explosion was carried out after a delay of about 10 min. 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 was controlled to an accuracy of 10 Pa (0.01% of composition). The flammable mixture was initiated by an electrical spark, giving 16 J energy for the explosion tests. For this study, three ignition positions were used, denoted as A, B and D, as shown in Fig. 1. The ignition position at C is not considered, as it will be discussed in our next paper. Repeatability and accuracy were confirmed by conducting at least three tests at each concentration.

3. Results and discussion

Previous studies indicate that pipe configurations and initial conditions have a significant impact on flame reactivity and the acceleration of hydrogen/air explosions \cite{23–26}. However, the effect of inhibitors on the flame reactivity of hydrogen/air mixtures in a pipe has not been fully explored. The discussion here will therefore focus on the pressure and flame-propagation mechanisms of hydrogen-inhibitors/air mixtures explosions in a tee-pipe configuration with the effect of different ignition positions.

3.1. Effect of inhibitors on pressure and flame speed during explosion development

Sets of recorded maximum overpressure along the tee pipe are given in Fig. 2 at three different ignition positions, denoted as A, B and D (see Table 1). These are in reference to the stoichiometric hydrogen/air mixtures, which were diluted with various amounts of CO₂, Ar and N₂. Flame speed and rate of pressure rise (dp/dt) are plotted in Figs. 3 and 4 as a function of pressure transducer points located along the pipe. Considering hydrogen as a prime fuel, a significant pressure drop of up to ∼50% and flame linearity in the presence of CO₂ were observed, particularly with the mixtures 95% H₂–5% CO₂/air, 95% H₂–2.5% CO₂–2.5% N₂/air and 95% H₂–2.5% Ar–2.5% CO₂/air. As reported by Di Benedetto et al. \cite{16}, the presence of free radicals, including O, H, CO, HCO, OH, HO₂, and CO₂, causes the rate of overpressure, flame speed and rate of pressure rise to decrease significantly, as illustrated in Figs. 2–4. Among the inhibitors, argon in the hydrogen/air mixtures gave a higher overpressure than the others. Movileanu et al. \cite{27} showed that the presence of additives changes the thermo-physical properties of the unburned mixtures in terms of thermal conductivity and heat capacity, and this condition will directly affect the overall flame reaction rate within the reaction zone. Kwon and Faeth \cite{13} also claimed that, even though nitrogen and argon have very similar transport properties, the addition of argon into hydrogen could lead to a higher laminar burning velocity due to increased flame temperature. This mechanism suggests that the effectiveness of inhibitors should be in the order of Ar < N₂ < CO₂.

On the other hand, we can postulate from Fig. 3 that there was a significant change in the flame speed of fuel–inhibitors/air mixtures along the centreline of the pipe and at the end points at all ignition positions. It can be said that fuels’ reactivity plays a major role within this context \cite{28}. Fig. 3 shows that the highest flame speed was at an average of 736, 719, 706 and 703 m/s for 95% H₂–2.5% Ar–2.5% N₂/air, 95% H₂–5% N₂/air, H₂/air and 95% H₂–5% Ar/air mixtures, respectively, for all ignition positions. These results are in good agreement with a previous study carried out by Kwon and Faeth \cite{13,14}. Comparing the flame speed of these mixtures with 95% H₂–5% CO₂/air under similar conditions, the linearity of the latter mixtures was higher, as the average recorded maximum flame speed was 599 m/s for all ignition positions. This shows that the addition of CO₂ into hydrogen mixtures caused the heat release rate to decrease, consequently lowering the average value of the flame speed. This present observation is in similar agreement with previous investigations in terms of the effects of CO₂ on reducing the flame propagation, thus decreasing the overall overpressure inside the pipe \cite{16}.

In order to further justify the effect of inhibitors on the explosion development of stoichiometric hydrogen/air mixtures, Lewis numbers were calculated as a basic analysis of the diffusivity rate of involved species in the unburnt gases, corresponding to the mass burning rate. This is the most important factor that affects the burning velocity and heat release rate during flame propagation \cite{27}. Lewis number, Le, was plotted as a function of pressure transducers’ points along the pipe as shown in Fig. 5. From the

**Table 1**

Position of each pressure transducer and thermocouple from ignition points.

<table>
<thead>
<tr>
<th>Ignition position</th>
<th>A</th>
<th>B</th>
<th>D</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Pressure transducers</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>P1</td>
<td>0.26</td>
<td>3.99</td>
<td>4.45</td>
</tr>
<tr>
<td>P2</td>
<td>1.18</td>
<td>3.07</td>
<td>3.53</td>
</tr>
<tr>
<td>P3</td>
<td>2.53</td>
<td>1.72</td>
<td>2.18</td>
</tr>
<tr>
<td>P4</td>
<td>3.42</td>
<td>0.83</td>
<td>1.29</td>
</tr>
<tr>
<td>P5</td>
<td>3.9</td>
<td>0.35</td>
<td>1.77</td>
</tr>
<tr>
<td>P6</td>
<td>4.03</td>
<td>1.44</td>
<td>0.6</td>
</tr>
<tr>
<td>P7</td>
<td>4.52</td>
<td>1.93</td>
<td>0.19</td>
</tr>
<tr>
<td><strong>Thermocouples</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>T1</td>
<td>0.45</td>
<td>3.84</td>
<td>4.25</td>
</tr>
<tr>
<td>T2</td>
<td>1.26</td>
<td>3.03</td>
<td>3.44</td>
</tr>
<tr>
<td>T3</td>
<td>1.65</td>
<td>2.64</td>
<td>3.05</td>
</tr>
<tr>
<td>T4</td>
<td>2.75</td>
<td>1.54</td>
<td>1.95</td>
</tr>
<tr>
<td>T5</td>
<td>3.42</td>
<td>0.87</td>
<td>1.23</td>
</tr>
<tr>
<td>T6</td>
<td>3.99</td>
<td>0.26</td>
<td>1.85</td>
</tr>
<tr>
<td>T7</td>
<td>4.12</td>
<td>1.54</td>
<td>0.56</td>
</tr>
<tr>
<td>T8</td>
<td>4.38</td>
<td>1.8</td>
<td>0.3</td>
</tr>
</tbody>
</table>
Fig. 2. Maximum overpressure vs. the recorded points in a tee pipe at different ignition positions.

Fig. 3. Maximum flame speeds vs. the pressure transducer points in a tee pipe at different ignition positions.
Fig. 4. Lewis number vs. the recorded points in a tee pipe at different ignition points.

Fig. 5. Rate of pressure rise vs. the recorded points in a tee pipe at different ignition positions.
figures, the value of the Lewis number for each of the fuel–inhibitors/air mixtures was lower than unity, which is in fair agreement with previous research data at NTP condition [29–31]. Analysis of the results presented in Fig. 5 clearly implies that all of the mixtures were affected by mass diffusivity, and that condition was more apparent when argon was added. The fact that the diffusion coefficient under normal conditions for Ar 

\[ D = \frac{k}{\rho C_p} \]  

(2)

where \( k \) is thermal conductivity (W/(m K)), \( \rho \) is density (kg/m³) and \( C_p \) is specific heat capacity ((J/(kg K))). The mass diffusivity dependence of the diffusion coefficient on temperature for gases for the current system can be expressed using:

\[ D = \frac{1.858 \times 10^{-3} T^{3/2}}{P} \sqrt{\frac{1}{M} + \frac{2}{R T}} \]  

(3)

Subscript 1 and 2 indicate the two different species present in the gaseous mixture; \( T \) is the absolute temperature (K), \( M \) is the molar mass (g/mol) and \( P \) is the relative pressure referring to \( \left( \frac{P}{P_r} \right) \) where \( P_r \) is the pressure on each measured point in this work [35].

The Lewis number effect can be further discussed based on the following observation: the flame propagation of the mixtures, in particular of the 5% N₂–95% H₂/air and 2.5% Ar–2.5% N₂–95% H₂/air mixtures along the pipe, was inconsistent with that of other mixtures when the mixtures ignited at position B, as illustrated in Figs. 3 and 5. The appearance of the multiple peak pressures (displayed in Fig. 6) indicates that, at a shorter obstacle distance, the flame experienced a strong interaction of transverse pressure waves from the tee junction and induced more turbulence. A recorded Lewis number of ~0.03 at the tee junction (P4) for those mixtures mentioned above suggests that the wrinkling of the flame became so great that the pockets of trapped unburned gas along the Tee junction area preheated. The net effect of the flame wrinkling and rapid turbulence is that the mass-burning rate of the flame increases, causing the flame to accelerate rapidly. This, subsequently, gives a higher overpressure. A similar situation was observed by Kim et al. [34], who demonstrated that the flame for a Le < 1 mixture was wrinkled by diffusion–thermal instability, accelerating the flame speed and consequently increasing the overpressure with time.

To justify the findings, the unburnt gas velocity ahead of the flame (\( S_g \)) was calculated and plotted for all the mixture compositions, as given in Fig. 7. As mentioned earlier, the flame reactivity of 5% Ar–95% H₂/air, and partially 5% N₂–95% H₂/air and 2.5% Ar–2.5% N₂–95% H₂/air, were mostly affected due to the dynamic effect, i.e., mass diffusivity and rapid mixing of the induced turbulence and faster flame downstream in the pipe. Since the induced maximum value of \( S_g \) was calculated to be more than 340 m/s, the flame travels at a greater speed, giving a massive compression impact at the
Fig. 6. Blast wave in P1, tee point, P5 and P7.
end pipe and thus, promoting a strong interaction between the fast flame and reflective wave. This phenomenon is known as retonation, and was significant when the ignition initiated at position A and D. Previous studies [15,16] provide similar observations on this phenomenon. However, the ignition position B showed the opposite impact on both pressure development and flame propagation in the tee pipe configuration, and retonation was barely observed. It can be interpreted that the blended inhibitors–fuel/air mixtures did not guarantee that the explosion risk posed on the obstacles would be minimised. Instead, in this study, when inerting was considered and ignition occurred at B, a ‘vulnerable condition’ resulted. As shown in Fig. 2, the maximum overpressures for 2.5% CO2–2.5% N2–95% H2/air, 2.5% Ar–2.5% N2–95% H2/air and 2.5% CO2–2.5% Ar–95% H2/air were observed at P2 when ignited at B. Previous research has shown that, with a wider fuel variation, i.e., composition of the blended mixtures, the Lewis number is directly proportional to the wider Markstein numbers [36]. This implies that the flame acceleration, with ignition at position B, was mainly affected by mass diffusivity (refer to Fig. 5), as mentioned in the previous section. It further suggests that the transition of the flame deformation into detonation and a strong interaction of reflective waves and fast flames downstream of the pipe could be possible causes of the maximum overpressure at P2. Furthermore, at ignition position B, the mixtures consisting of 5% N2–95% H2/air and 2.5% Ar–2.5% N2–95% H2/air gave the highest overpressure, recorded at P5 and P7 in the former and P5 for the latter. However, it can be postulated that one of the dominant causes of the fast flame propagation was mass diffusivity, particularly in the presence of Ar and N2, as explained in the previous section. This phenomenon is also discussed by Qiao et al. [14].

3.3. Explosion severity

Based on the rate of pressure rise, it can be depicted that the severity of all of the mixtures at ignition point B was higher than the others. Moreover, the maximum rate of pressure rise values were measured at 78.08 ¥ 105, 74.64 ¥ 105, 73.82 ¥ 105 and 58 ¥ 105 Pa s⁻¹ at all of the ignition points for 95% H2–2.5% Ar–2.5% N2/air, 95% H2–5% N2/air, H2/air and 95% H2–5% Ar/air, respectively (refer to Fig. 4). From the data, it is found that flame mixtures with CO2 had lower severity. This suggests that severity in the pipeline not only depends on the vessel size [37], initial condition and mixtures [10], but also on points of ignition.

4. Conclusion

Based on the importance of hydrogen fuel for future transportation, this research was conducted to investigate the effectiveness of argon, nitrogen and carbon dioxide on hydrogen/air flame acceleration in a tee pipe configuration.

The findings can be summarized as follows:

1. The ignition position and initial fuel composition had a strong influence on the overpressure, flame speed and rate of pressure rise. The blending of the inhibitors gave a lower severity in all the studied compositions of hydrogen/inhibitors/air mixtures. From the study, CO2 is the most efficient inhibitor, followed by N2 and argon.

2. The explosion severity recorded at ignition position B was higher than at A or D. Flame acceleration, with ignition at position B, was mainly affected by mass diffusivity; consequently, it can be said to promote the transition of flame wrinkling, which increases the mass burning rate in the system. However, flame acceleration at ignition positions A and D was affected by the
strong interaction of a reflective wave at the end pipe and a fast induced flame, highlighting the retonation-like obstacles phenomenon.

3. Of the studied mixtures, 95% $\text{H}_2$–2.5% $\text{Ar}$–2.5% $\text{N}_2$/air, 95% $\text{H}_2$–5% $\text{N}_2$/air, $\text{H}_2$/air and 95% $\text{H}_2$–5% $\text{Ar}$/air showed the highest severity risks in terms of rate of pressure rise. This is due to their dynamic properties. However, it was found that the mixtures in the presence of $\text{CO}_2$ had lower severity, since the average recorded maximum flame speed was lower at all of the ignition points (at an average of 599 m/s).

Most important to clarify are the methodologies to classify the propagating flames on a tee pipe configuration, taking into account the distance position of the tee pipe junction as a function of the ignition position. This phenomenon will form the focus of our next paper.

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