Self-ignition of hydrogen-nitrogen mixtures during high-pressure release into air

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Presentation plan

1. Introduction
2. Experimental stand and procedure
3. Results
4. Numerical simulations
5. Results
6. Conclusions
1. Introduction

**Hydrogen ignition without distinct ignition source:**
- 1922 - Anon
- 1930 - Fenning and Cotton
- 1965 - Reider et al.
- 1972 - Wolanski and Wojcicki, ‘diffusive ignition model’

Recent research in hydrogen ‘self-ignition’ process:
- Pure hydrogen release,
- Various geometrical configurations and boundary conditions:
  - Extension channel length
  - Extension channel cross-section shape (round, rectangle)
  - Extension channel material (metal, glass)
  - Diaphragm: material, opening rate, initial curvature
  - Obstacle presence in the front of the hydrogen stream
- Shorter tubes <40 mm lead to significant increase in $P_{\text{burst}}$
- High discrepancies observed due to the different experimental stands and procedures utilized:
  - fast valves/slow filling
  - diaphragm burst/‘tulip shape’ opened/punctured

*Little data regarding gas doping influence on self-ignition process.*
2. Experimental stand and procedure

Control and data acquisition section: computer, amplifier, time unit, PCB pressure sensors, ion probes, ionisation probes

Air section
1x0.11x0.11 m tube with optical access, diaphragm holder and extension tube

High-pressure section
gas cylinders, single acting gas-driven booster – Haskel AG-75, high-pressure line, pressure accumulator, fast acting electromagnetic valve

Diaphragms made of aluminium,
Thickness: 0.1, 0.15, 0.2, 0.25 mm

Sensors positions regarding diaphragm and flow direction (PS – pressure sensors, IP – ion probes, PD – photodiodes):
• PS1 – 30 mm upstream,
• PS2 – 60 mm downstream,
• PS3 – 860 mm downstream,
• IP1 – 360 mm downstream,
• IP2 – 560 mm downstream,
• PD1 – 100 mm downstream,
• PD2 – 125 mm downstream,
• PD3 – 150 mm downstream.
2. Experimental stand and procedure cont.

![Experimental setup with different diameters: d = 6 mm, d = 10 mm, d = 14 mm.](image)
2. Experimental stand and procedure cont.

30 ms signal

diaphragm ruptures at Plane

gas depressurizes into the air through tube

EMV

vacuum pump connection

pressure accumulator

vent connection

PS1

extension tube: d = 6, 10, 14 mm, L = 10 – 100 mm

EMV

diaphragm

psp

ambient air

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3. Results

Total number of experiments 655:
$\text{H}_2 - 360$
$\text{H}_2+\text{N}_2$: 
5% $\text{N}_2$ - 213 
10% $\text{N}_2$ - 82

Extension tubes:
$\text{d} = 6, 10 \text{ and } 14 \text{ mm}$,
$\text{L} = 10, 25, 40, 50, 75, 100 \text{ mm}$,

Initial pressure range
2 - 18 Mpa

Initial temperature: 
ambient $\sim 298 \text{ K}$

3. Results cont.

With ignition

\[ P_{\text{burst}} = 9.76 \text{ MPa} \]

- pressure drop in PS1 sensor – diaphragm burst
- clear photodiodes signals when gas leaving the tube – ignition in extension tube
- ionisation probes signals \( \rightarrow \) sustained combustion in the further part of the tube
- pressure sensors PS2 and PS3 higher indications due to the combustion
- PS2 and PS3 oscillations – shock reflections

Without ignition

\[ P_{\text{burst}} = 9.77 \text{ MPa} \]
3. Results cont.

100% H₂

- Self-ignition is possible only above certain burst pressure
- ‘Ignition limit’ – minimum initial pressure for which ignition was observed
- Ignition limit decreases with extension channel length increase
- No ignition for extension channels with L < 40 mm → non-linear dependence observed previously
3. Results cont.

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### 3. Results cont.

**H₂ + N₂**

![Graph showing the relationship between Pₜₐₐ₉ (MPa) and L [mm] for various d (diameter) values and different N₂ concentrations (5% and 10%) for H₂ + N₂.]

<table>
<thead>
<tr>
<th>Pₜₐ₉ mm</th>
<th>d = 6 mm</th>
<th>10 mm</th>
<th>14 mm</th>
<th>d = 6 mm</th>
<th>10 mm</th>
<th>14 mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>100 mm</td>
<td>1.4</td>
<td>2.08</td>
<td>2.12</td>
<td>1.87</td>
<td>2.85</td>
<td>2.72</td>
</tr>
<tr>
<td>75 mm</td>
<td>1.25</td>
<td>1.21</td>
<td>1.33</td>
<td>1.83</td>
<td>1.83</td>
<td>1.81</td>
</tr>
<tr>
<td>50 mm</td>
<td>1.19</td>
<td>1.39</td>
<td>1.15</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>40 mm</td>
<td>1.14</td>
<td>1.34</td>
<td>1.15</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

**H₂ + CH₄**

![Graph showing the relationship between Pₜₐ₉ (MPa) and L [mm] for various d (diameter) values and different CH₄ concentrations (5% and 10%) for H₂ + CH₄.]

3. Results cont.

**Ignition limit points compared to the postshock conditions (Cantera calculations)**

Postshock $P_2$, $T_2$ as a function of driver gas composition and pressure

Results scatter shows that ideal shock assumptions do not include real 3-dimensional effects of the release including shock-shock, shock–wall interactions and diaphragm opening process.

Higher postshock conditions required for ignition in shorted tubes ($L= 40-50$ mm)

None of the results is below the autoignition temperature of hydrogen-air mixture ($T= 858$ K).
4. Numerical simulations

Kiva3V
- ALE methodology
- Code substantially modified by Wen and co-workers, details in:
  Xu et al. J.Loss Prev. Proc. Ind. 21, 2008 and 22, 2009,
  Wen et al. Comb. Flame 156, 2009,
  Wen et al. ICHS 2013, Brussels, Belgium

- Selected part of experimental stand geometry
- Axisymmetrical domain
- Saxena and Williams’s reaction model (21 reactions)
- 1 200 000 cells
- Structural mesh in extension tube - 15 μm
- Orthogonal mesh in high-pressure part of the domain 15-150 μm
- Diaphragm 0.1 mm thick, opening time 5 μs

<table>
<thead>
<tr>
<th>$P_{\text{burst}}$ [MPa]</th>
<th>100% $\text{H}_2$</th>
<th>5% $\text{N}_2$</th>
<th>10% $\text{N}_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>X</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>X</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>X</td>
<td>X</td>
<td>X</td>
</tr>
<tr>
<td>10</td>
<td>X</td>
<td>X</td>
<td>X</td>
</tr>
<tr>
<td>12</td>
<td></td>
<td></td>
<td>X</td>
</tr>
</tbody>
</table>
5. Results

100% H₂, $P_{\text{burst}} = 6$ MPa, frames every 5 μs starting from 1 μs of simulation time, first ignition near wall after 16 μs.
5. Results cont.

100% H₂ cases for contact surface position near tube end

- **P = 5 MPa, τ = 40 µs**
- **P = 6 MPa, τ = 40 µs**
- **P = 8 MPa, τ = 36 µs**
- **P = 10 MPa, τ = 34 µs**

Increase in the mixing intensity at the contact surface due to the stronger leading shock influence. Number of ignition spots:

- 5 MPa – 1 at the wall
- 6 MPa – 2 at the wall
- 8 MPa – 3: 2 at the wall, 1 in the channel axis
- 10 MPa – 3: 2 at the wall, 1 in the channel axis
5. Results cont.

Maximum temperatures recorded during simulations

- First temperature peak at 5µs time – diaphragm fully open,
- Progressive temperature increase due to the shock waves reflections
- Higher nitrogen addition and lower initial pressure delay first ignition event
5. Results cont.

Ignition and following sustained combustion requires at least 3 ignition spots. Reaction should spread to the whole contact surface before leaving the tube which is consistent with previously done research (Lee et al. 2011, Kim et al. 2013, Grune et al. 2013).
5. Results cont.
Diaphragm opening time influence

$V_{\text{shock}} \approx 1350 \text{ m/s}$  
$V_{\text{shock}} \approx 1315 \text{ m/s}$  
$V_{\text{shock}} \approx 1240 \text{ m/s}$  
$V_{\text{shock}} \approx 1084 \text{ m/s}$  
$V_{\text{shock}} \approx 890 \text{ m/s}$

$P_{\text{burst}} = 6 \text{ MPa}, \ 100\% \ H_2$

0 µs – ideal case, shock velocity close to theoretical value

- finite opening process provides shock waves reflections which enhance mixing due to the instabilities generated at the multi-shocked contact surface
- Increase in opening time influences initial shock wave structure and initial shock velocity
6. Conclusions

• Experimental and numerical research on H₂ and H₂-N₂ mixtures self-ignition process during release into air was presented
• In total, 295 experiments were conducted for H₂+N₂ mixtures (+360 for pure H₂)
• Self-ignition process highly depends on the extension tube length, initial pressure and mixture composition
• N₂ addition considerably increases the mixture’s initial pressure necessary for the self-ignition to occur, effect is stronger for longer tubes and may increase pressure as much as 2.12 or 2.85-times for 5% and 10% of N₂ in the mixture with H₂ respectively

• KIVA-3V simulations showed that depending on the initial pressure of the mixture, specific ignition sequence is present with 1, 2 or 3 ignition spots at various locations
• 1st and 2nd ignition spots are present in the vicinity of the extension channel wall
• 3rd ignition spot is present at the tip region of the contact surface
• N₂ addition ‘shifts’ the initial pressure necessary for the ignition sequence to occur
• Comparison with experimental results pointed at condition for 3 ignition spots presence for sustained combustion after leaving the tube
• Diaphragm opening time highly influences initial shocks structure and shock velocity, thus instabilities generated at the contact surface
Acknowledgements

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Thank you for your attention!
References