Analysis of Vortex Tube Applications in Hydrogen Liquefaction

Marshall Crenshaw

Washington State University
Pullman, WA
November 14, 2017
Why is Renewable Energy Necessary?

- >80% of global energy comes from fossil fuels.¹
- Since 1976, global temperatures warmer than long-term average.²
- 11 of the top 12 hottest years are from 2003-2016.³

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² https://climate.nasa.gov/scientific-consensus/
³ https://www.ncdc.noaa.gov/sotc/global/201613
H₂ Production

• 9th most abundant element in earth’s crust
• Photo-biological bacteria produce hydrogen

Difficulties of H₂

- Lowest volumetric density
- Containers are pressurized or cryogenic to increase density

https://energy.gov/eere/fuelcells/hydrogen-storage
Liquid vs Pressurized H₂

- Double density when liquefying over pressurizing
Diagram of Vortex Tube (VT)

- Inexpensive cooling device
- 1 high pressure stream $\rightarrow$ 2 low pressure streams
- Transfers cold stream energy $\rightarrow$ hot outer stream
- No moving parts
Pre-cooled Linde-Hampson Cycle

- Low-cost cycle for hydrogen liquefaction
- Low-efficiency method for liquefaction
- Increase cycle efficiency by adding component
Pre-cooled L-H cycle with VT

- Additional compressor and heat exchanger
- Cycle efficiency: Adding compressor > Adding throttle
- 3rd HX reduces O-P ratio of hydrogen
- What is O-P ratio?
Orthohydrogen & Parahydrogen

- Equilibrium O-P ratio depends on temperature of hydrogen
- Normal hydrogen $\rightarrow$ 3:1 O-P ratio
- Liquid hydrogen $\rightarrow$ all parahydrogen
- Catalyst added to HXs to maintain equilibrium O-P ratio
- Why is equilibrium hydrogen important?
Importance of Ortho-Para Equilibrium

• 50% of normal hydrogen boils away in 200 hours

Statistical Thermodynamic Properties of $\text{H}_2$
Stat Thermo

https://hydrogen.wsu.edu/2015/06/22/why-equilibrium-hydrogen-doesnt-exist/
REFPROP

Ideal-gas enthalpy [MJ/kg] vs Temperature [K]

- Ortho [REFPROP]
- Para [REFPROP]
REFPROP with Corrected Orthohydrogen vs. Stat Thermo

![Graph showing comparison of enthalpies and temperatures for different hydrogen states.]

- Ideal gas enthalpy [MJ/kg]
- Conversion enthalpy [MJ/kg]
- Temperature [K]

Corrected Ortho
Para [REFPROP]
Stat Thermo
MATLAB

$r^2 = 0.999996$

https://hydrogen.wsu.edu/2015/06/22/why-equilibrium-hydrogen-doesnt-exist/
Equilibrium Orthohydrogen Fraction

Temperature (K)

Equilibrium Orthohydrogen Fraction
Piece-Wise Equilibrium H₂ Function

\[ O_{frac} = \begin{cases} 
    a_1 T^2 + a_2 T + a_3 & T \leq 24.82K \\
    b_1 e^{b_2 T} + b_3 & T \leq 51.51K \\
    \sum_{i=1}^{4} c_i T^i + c_5 & T > 51.51K 
\end{cases} \]

| Coefficients of equilibrium MATLAB function |
|---|---|---|
| \( a_1 \) | 1.935x10⁻⁴ | \( b_1 \) | -2.044 | \( c_1 \) | 1.111x10⁻⁷ |
| \( a_2 \) | -5.832x10⁻³ | \( b_2 \) | -2.724x10⁻² | \( c_2 \) | -2.243x10⁻⁶ |
| \( a_3 \) | 3.448x10⁻² | \( b_3 \) | 0.75 | \( c_3 \) | -1.886x10⁻⁵ |
| \( a_4 \) |  | \( b_4 \) |  | \( c_4 \) | 6.514x10⁻⁴ |
| \( \sum_{i=1}^{4} c_i T^i + c_5 \) | | | | \( c_5 \) | -3.499x10⁻³ |
Approximate Equilibrium O-P Function

$\text{Equilibrium Orthohydrogen Fraction}$

$r^2 = 0.99986$

- Stat Thermo
- MATLAB
Liquefaction Components & Cycle Assumptions
LN2 Bath

• Bath pressure is atmospheric (101.3 kPa) and temperature is 77 K
• LN$_2$ Bath is catalyzed: Ofrac = \(~0.5\)

• Heat extracted:

\[ \dot{Q}_{Bath} = \dot{m}_{H_2}(h_{Bath,in} - h_{Bath,\text{out}} + \Delta h_{Bath,\text{conv}}) \]

• Added work to hydrogen liquefaction:

\[ \dot{W}_{Bath} = \frac{\dot{Q}_{Bath} \left( \frac{LN_2\text{ideal}}{LN_2FOM} \right)}{\dot{m}_{LH_2} \cdot \Delta H_{vap,N_2}} \]
Counter-flow Heat Exchanger

- Energy balance equation:

$$\sum_{i=1}^{n} (\Delta h_i + \Delta h_{\text{conv},i}) = 0$$

- Minimum temperature approach solves for outlet temperatures

- Hot stream T limit: $$T_{\text{Hot, out, limit}} = \Delta T_{\text{min}} + T_{\text{Coldest, in}}$$

- Cold streams T limit: $$T_{\text{Cold, out, limit}} = T_{\text{Hot, in}} - \Delta T_{\text{min}}$$

Diagram:

- Cold
- Hot
- Nitrogen

HX2, HX3, HX4
Staged Compression with Intercooling

- Staging compressors decreases their work
- 4 stages is optimal for compressors
Compressor/Expander Component

- Pressure Ratio per stage: $PR_{\text{stage}} = \sqrt[n]{\frac{P_{\text{out}}}{P_{\text{in}}}}$

- Enthalpy of compressor: $h_2 = \frac{h_s - h_1}{\eta_C} + h_1$

- Work of compressor: $N_C = \sum_{i=1}^{n} \dot{m}_C \left( h_{2,i} - h_{1,i} \right)$

- Heat extracted: $\dot{Q}_C = N_C + \sum_{i=1}^{n} \left( h_i - h_{i-1} \right)$

- Enthalpy of expander: $h_2 = \eta_E (h_1 - h_s) + h_1$

\[\begin{align*}
\text{C} & \quad \uparrow \quad \dot{Q}_C \\
N_C & \downarrow \\
\text{E} & \downarrow \\
N_E & \uparrow
\end{align*}\]
Throttle & Tank

• Throttle is ideally isenthalpic: \( h_{in} = h_{out} \)

• Quality of \( \text{LH}_2 \): solved with enthalpy & pressure

• Heat of O-P conversion in tank:
  \[
  \dot{Q}_{\text{Tank,conv}} = m_{\text{Tank,in}} \Delta h_{\text{conv}}
  \]

• Return stream flow:
  \[
  \dot{m}_{\text{Tank, out}} = \dot{m}_{\text{Tank,in}} \cdot X_{\text{H}_2} + \frac{\dot{Q}_{\text{Tank, conv}} + \dot{Q}_{\text{Tank, heat leak}}}{h_{\text{LH,H}_2}}
  \]

\[ JT \]

Liquid \( \text{H}_2 \) Tank
Effectiveness of a VT

- Specific exergy: $X_1 = h_0 - h_1 - T_0(s_0 - s_1)$

- Exergy destruction:
  $$\Delta X_{Dest} = \sum (m_{in}X_{in}) - \sum (m_{out}X_{out}) - W$$

- Limits are set to the exergy destruction of throttle (lower) and expander (higher)
Effectiveness of a VT cont.

- Exergy destruction of devices:

\[
\Delta X_{VT} = \dot{m}_{VT,\text{in}}X_{VT,\text{in}} - \dot{m}_{VT,\text{cold}}X_{VT,\text{cold}} - \dot{m}_{VT,\text{hot}}X_{VT,\text{hot}}
\]

\[
\Delta X_{JT} = \dot{m}_{VT,\text{cold}}(X_{VT,\text{in}} - X_{JT,\text{cold}}) + \dot{m}_{VT,\text{hot}}(X_{VT,\text{in}} - X_{JT,\text{hot}})
\]

\[
\Delta X_{Exp}
\]

\[
= \dot{m}_{VT,\text{cold}}(X_{VT,\text{in}} - X_{Exp,\text{cold}}) - W_{Exp,\text{cold}} + \dot{m}_{VT,\text{hot}}(X_{VT,\text{in}} - X_{Exp,\text{hot}}) - W_{Exp,\text{hot}}
\]

- Exergy destruction using effectiveness

\[
\Delta X_{VT} = \Delta X_{Exp} + (1 - \varepsilon_{VT})(\Delta X_{JT} - \Delta X_{Exp})
\]
Variables of the VT

- Cold fraction: $VT_{cf} = \frac{\dot{m}_{VT,cold}}{\dot{m}_{VT,in}}$

- Pressure Ratio: $VT_{PR} = \frac{P_{in}}{P_{VT,cold}}$

- Hot outlet Pressure: $P_{VT,hot} = (\frac{1}{3})P_{VT,in} + (\frac{2}{3})P_{VT,cold}$

- Outlet temperatures are solved with effectiveness
## Cycle Parameters

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
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<tbody>
<tr>
<td><strong>Capacity</strong></td>
<td>1 TPD&lt;sup&gt;a&lt;/sup&gt;</td>
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<td><strong>Supply</strong></td>
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<tr>
<td>Pressure</td>
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<td>Temperature</td>
<td>300 K</td>
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<td><strong>Dead State</strong></td>
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<tr>
<td>Pressure</td>
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<tr>
<td>Temperature</td>
<td>300 K</td>
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<td><strong>Compressor</strong></td>
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<td>Efficiency</td>
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<td>Peak Pressure</td>
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<td>Stages</td>
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<td>Pressure ratio (per stage)</td>
<td>3.16&lt;sup&gt;b&lt;/sup&gt;</td>
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<td><strong>Heat Exchanger</strong></td>
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<tr>
<td>Pinch point ΔT</td>
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<tr>
<td>Ortho-Para conversion</td>
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<td><strong>Refrigerant</strong></td>
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<td>LN&lt;sub&gt;2&lt;/sub&gt;</td>
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<td>Precoolant Figure of Merit</td>
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<td>H&lt;sub&gt;2&lt;/sub&gt;</td>
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<td>20.2 K</td>
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<td>Ortho Fraction</td>
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<sup>a</sup> Tonne per day  
<sup>b</sup> Value only pertains to L-H cycle; L-H w/VT cycle varies
## Previous Cycle Parameters

<table>
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<tr>
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<tr>
<td>Efficiency [%]</td>
<td>79[^a]</td>
<td>85[^b]; 80[^c]; 85[^d]; 85[^b]</td>
<td>-</td>
<td>80[^b]</td>
<td>65 – 70[^e]</td>
<td>80[^b]; 80[^b] [He]</td>
<td>85[^b]; 70[^f] [He/Ne]</td>
<td>92[^b] [He]</td>
<td>80[^b]</td>
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<td>Peak Pressure [MPa]</td>
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<td>79[^b]; 84[^g] [N₂]</td>
<td>85[^b]; 90[^h]; 85[^b]</td>
<td>-</td>
<td>80[^b]</td>
<td>&gt; 85[^b]</td>
<td>85[^b]; 83[^i]; 80[^b]; 86[^b]</td>
<td>80[^b]</td>
<td>93[^b]; 92[^b]; 90[^b]; 88[^b]</td>
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<td>96</td>
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<td>0.06 – 9[^i]</td>
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<td>LN₂</td>
<td>MR[^c]</td>
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<td>90[^b]</td>
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<tr>
<td>Pressure [MPa]</td>
<td>0.929</td>
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<td>0.13</td>
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<td>0.13</td>
<td>0.1</td>
<td>0.1</td>
<td>0.15</td>
<td>0.106</td>
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<tr>
<td>Ortho Fraction [%]</td>
<td>3</td>
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<td>&lt; 5</td>
<td>5</td>
<td>&lt; 5</td>
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<td>≤ 1</td>
<td>~0.2[^f]</td>
<td>≤ 5</td>
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<tr>
<td>Ideal [kWh/kg]</td>
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<td>2.86</td>
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<td>8.73</td>
<td>5 – 7[^g]</td>
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<td>47.15</td>
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<td>23.61</td>
<td>44.56</td>
<td>78.8 – 56.26[^h]</td>
<td>47.8</td>
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</table>

[^1]: Information acquired from Berstad et. al [49]
[^2]: Information acquired from Krasae-in et al [87]
[^a]: Weighted average
[^b]: Isentropic
[^c]: Mixed Refrigerant; composition varies by cycle
[^d]: Combined with a cold compressor
[^e]: Isenthalpic
[^f]: ΔT/ΔT = 0.03
[^g]: Dependent on Feed Pressure
[^h]: Polytropic
[^i]: Equilibrium composition assumed
[^j]: At 80 K, H₂ is converted to an $\eta_{rec} = 0.53$, the following is continuous
[^k]: Unknown
Cycle Analysis & Results
State-Point Analysis of Ideal Cycle

- Liquid hydrogen yield: $\varphi = \frac{\dot{m}_f}{\dot{m}}$

- Ratio of nitrogen to compressed hydrogen flow: $\psi = \frac{\dot{m}_{N_2}}{\dot{m}}$

- Liquid yield maximum: $\varphi_{max} = \frac{(h_9 - h_4)}{(h_9 - h_7)}$

- Relating $\psi$ to $\varphi_{max}$: $\psi = \frac{(h_1 - h_2)}{(h_{11} - h_{10})} + \varphi_{max} \frac{(h_7 - h_1)}{(h_{11} - h_{10})}$

O-P conversion at LH2 Tank

- O-P conversion happens at LN₂ Bath and LH₂ tank

- $m_{boil} = \frac{\dot{Q}_{Tank}}{h_g - h_f}$

- $m_\ast = m + \frac{m_{boil}}{\varphi_{max}}$

- $\varphi_\ast = \frac{m_f}{m_\ast} = \frac{\varphi_{max}}{1 + \frac{\dot{Q}_{Tank}/m_f(h_8 - h_7)}{m_f}}$

- $\psi_\ast = \frac{m_{N2}}{m_\ast}$

Diagram:

- $m_\ast - m_f$ to $m_\ast$
- $m - m_f$ to $m_\ast$
- $m_{boil}$ from $m_f$
- $m_f$ from $m_{boil}$
- $\dot{Q}_{Tank}$ to $m_{boil}$

Liquid H₂ Tank
Work of LN₂ Bath

- Maximum yield of LN₂ Bath: \( \varphi_{N_2,\text{max}} = \frac{\dot{m}_{N_2,f}}{\dot{m}_{N_2}} = \frac{(h_{N,1} - h_{N,2})}{(h_{N,1} - h_{N,5})} \)

- Work of LN₂ Bath: \( \alpha = \frac{1}{\varphi} \left( \frac{\dot{W}}{\dot{m}} \right)_{\text{comp}} \)

- Work of compressor: \( \left( \frac{\dot{W}}{\dot{m}} \right)_{\text{comp}} = T_{in} \cdot (s_{in} - s_{out}) - (h_{in} - h_{out}) \)

- \( \alpha_{N_2} = \frac{1}{\varphi_{N_2,\text{max}}} \left[ T_{N,1} \cdot (s_{N,1} - s_{N,2}) - (h_{N,1} - h_{N,2}) \right] \)
State-Point Results

• State-Point work of liquefaction for pre-cooled L-H cycle:

\[ \alpha_{H_2} = \frac{1}{\varphi_*} \left[ T_1 \cdot (s_1 - s_2) - (h_1 - h_2) \right] + \alpha_{N_2} \left( \psi_* + \frac{\dot{Q}_{\text{Bath}}}{h_{12} - h_{11}} \right) \]

• Peak pressure of LN2 Bath: 100 bar (10 MPa)

• Temperature of LN2 Bath: 63.2 K or 77 K

• State-Point work of liquefaction results:
  - \( \alpha_{s,63.2\,K} = 13.65 \text{ kWh}\cdot\text{kg}^{-1} \)
  - \( \alpha_{s,77\,K} = 19.95 \text{ kWh}\cdot\text{kg}^{-1} \)

• Peschka value: \( \alpha_P = 16.27 \text{ kWh}\cdot\text{kg}^{-1} \)
State-Point vs. Component Results

- Work of liquefaction for component-based model:
  - $\alpha_{c,63.2\,K} = 14.82\,\text{kWh}\cdot\text{kg}^{-1}$
  - $\alpha_{c,77\,K} = 20.83\,\text{kWh}\cdot\text{kg}^{-1}$

- Percentage error between results:
  - 63.2 K: 8.57%
  - 77 K: 4.41%

- Error caused by heat exchanger and LN$_2$ components
Vortex Tube Simulation

![Graph showing the relationship between effectiveness and temperature drop with pressure ratio.](image)
Reduced Work for L-H w/VT Cycle
Cycle Analysis & Results
Simulation of L-H w/VT Cycle
Recommendations

• Simulate higher complexity cycles e.g. pre-cooled Claude

• Vary the LN$_2$ Bath temperature to 63.2 K

• Higher inlet pressure tests for vortex tube

• Continued tests of vortex tube catalyzation

• Test vortex tube as a final liquefaction component
Summary

• Simplified Statistical thermodynamic models created
• Variable components created for the pre-cooled L-H cycle
• Component model verified with state-point model
• Effectiveness introduced as a new VT variable
• Required effectiveness provided for combinations of $VT_{cf}$ and $VT_{PR}$ values