Expansion vessel for supercritical hydrogen in a spallation neutron source moderator circuit

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Abstract. High-energy neutrons are being decelerated by passing through supercritical parahydrogen circulated by pumps in a closed loop. Fluctuations in neutron heat load cause changes of the circuits’ local and average temperature and hence significant pressure variations caused by the almost incompressible behavior of hydrogen. Solutions by adding a variable volume in form of a helium gas-backed metal bellow to mitigate pressure deviations are already in use. This paper presents an alternative approach by introducing a vertical storage vessel for supercritical hydrogen in a side branch of the moderator loop, with cold incompressible high density hydrogen at the bottom and warmer compressible lower density hydrogen at the top.

1. Introduction

In a spallation neutron source neutrons are being generated by accelerating protons which hit a metal target (e.g. tungsten) and set free neutrons of a broad energy spectrum due to the disintegration of the activated metal atoms. For the moderation process or the deceleration of high-energy neutrons supercritical parahydrogen is the preferred cooling fluid [1, 2] to supply cold neutrons to scattering experiments. The basic elements of a closed cooling circuit can be seen in figure 1.

Figure 1. Simplified flow scheme of a spallation neutron source moderator cooling circuit with supercritical parahydrogen (antiparallel proton spin).

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In a closed loop cryogenic hydrogen is circulated at temperatures around 17 K and pressures around 1.5 MPa to the moderator vessel where neutrons interact with parahydrogen molecules. The neutron energy reduction causes the main heat load into the fluid. A helium refrigerator has to recool the hydrogen. The circulator normally operates with a constant mass flow under all conditions with a nominal duty ensuring a temperature increase of less than or equal to 3 K over the moderator vessel at maximum neutron beam power. In the following a pressure drop of 0.14 MPa, a mass flow rate of 1 kg/s and a total loop volume of 300 liter is assumed. The static heat load causes a 0.5 K temperature increase (beam OFF = no neutrons). A dynamic heat load at full beam power leads to an additional temperature increase of 3 K (beam ON = max. number of neutrons). Pressures above the critical point for parahydrogen of 1.28 MPa are required in the moderator vessels at all times to prevent boiling near the significantly more neutron-heated aluminum walls. A remarkable feature of hydrogen with the above described temperature and pressure range is the nearly incompressible behavior as shown in figure 2. An isochoric average temperature increase at 1.5 MPa from 17.25 to 18.75 K results in an intolerable high pressure increase over the design pressure. Starting from average 18.75 K cooling down to 17.25 K would lead to boiling and subatmospheric pressure. This pressure fluctuation due to planned or sudden changes like instant beam trips has to be mitigated to stay in allowable boundaries.

![Figure 2. Phase diagram of parahydrogen with intermediate loop temperature and pressure under neutron beam OFF (cross) / beam ON (rectangle) conditions and the two corresponding isochoric lines.](image)

2. Pressure Control Systems in Moderator Loops Circulating Supercritical Hydrogen.
A changed neutron heat load or average temperature and implications on pressure can be mitigated by three mechanisms:

- keep the average temperature constant by changing temperature distribution (variation helium refrigerator cooling power and/or electrical heating in the hydrogen loop),
- change of loop volume by approximately 1.4 % (see figure 3),
- change of mass in the main loop by approximately 1.4 % (see figure 4 and 5).
2.1. Pressure control by heating and/or cooling

The pressure can be kept stable when the average loop temperature is kept almost constant for different neutron heat loads. An electrical heater could substitute for a missing neutron heat load. Placing the heater close to the moderator is impossible due to very limited space and irradiation. Depending on the location of the heater it can substitute either for a minor impact on the average temperature (far downstream of the moderators) or the moderator inlet temperature would be increased for a short time (installation after the heat exchanger) when the beam power would be increased again. Another approach is the adjustment of cooling power of the heat exchanger. The helium mass flow can be controlled with a bypass and thereby change the cooling power. This again would raise the inlet temperature of the moderator for a returning beam for a short time. The heater downstream of the moderator and refrigeration power control together could be combined. Depending on the allowed pressure variation such a system would have to react within a few seconds which is challenging.

2.2. Pressure control by variable volume

A widely used alternative solution is the application of immersed helium gas filled metal bellows that will perform an expansion or contraction of the gas and bellows enabling the necessary change of specific volume for a stable pressure. The metal bellows is connected to a warm gas storage to offer a larger expansion volume at higher temperature and compressibility and thus smaller pressure difference due to variation in neutron beam power. This technique is in use at a couple of facilities [1, 2] but has also shown difficulties with earlier development stages.

2.3. Pressure control by variation of mass

The third method of mitigating pressure variations due to a change in average loop temperature is to remove or introduce mass to the loop and thereby adjust the specific volume. This hydrogen has to be stored in a buffer vessel in a side branch of the circuit. This storage vessel could be either at ambient temperature or at a temperature close to the loop depending on the position of the helium refrigerator. An ambient vessel would be possible if the helium refrigerator is close to the hydrogen circulation and the helium heat exchangers could be used cooling down or warming up the small hydrogen flow. This option is not being evaluated in the following. Alternatively, it will be focused on a cold storage vessel. This approach has already been proposed earlier for a supercritical helium circulation in a fusion reactor [5]. The buffer is a vertical cylindrical vessel which is rather long and small in diameter. At the bottom it is connected to the pump discharge side without a valve and it contains a small amount of hydrogen slightly above the loop temperature. On top and after a temperature transition zone a larger volume of 40 K hydrogen with seven times less density is arranged. Since operation is above critical pressure, no distinct liquid-gas boundary exists but a rather smooth change in density. The top of the vessel is connected to the suction side of the pump, controlled by a valve guiding any leaving fluid through a heat exchanger cooled by the helium refrigerator flow. Hydrogen can enter only through the bottom but leaves the vessel through the top (open valve) or the bottom (heat and expand the 40 K volume with closed valve). The temperature gradient has to be maintained by continuously heating of the top and cooling of the bottom. Furthermore, a small flow through the bottom and out of the top is required to keep the temperature distribution constant. Two different operating methods can be chosen: Passive or actively controlled buffer vessel.

2.3.1. Passive cold storage vessel. An increase from no to full neutron beam power leads to a rising average loop temperature and specific density. As can be seen in figure 6, cold fluid will leave the circuit entering the storage vessel at the bottom. The valve at the top stays closed. The 40 K vapor at the top will then be compressed isentropically from 1.32 MPa to the nominal operating pressure of 1.57 MPa at the pump discharge due to the rising liquid piston. If the beam power is being decreased the 40 K vapor will expand isentropically and push cold liquid from the bottom of the vessel back into the loop. For the following calculations an overall buffer volume of 60 liter including 3 liter at
17.25 K, 6 liter temperature transition zone from 17.25 to 40 K (both set as incompressible) and 51 liter at 40 K have been assumed.

![Diagram of passive cold storage vessel](image)

**Figure 6.** Passive cold storage vessel (exemplary for a total volume of 60 liter) with transition from beam OFF (left) to beam ON (right) and accompanied isentropic compression.

The change of loop liquid volume equals the necessary negative change of 40 K vapor in the buffer vessel for an isentropic compression as shown in equation (1). For a defined mean temperature and allowable pressure change in the main loop the derivatives of specific volume over pressure for constant temperature and specific volume over temperature for constant pressure (see table 1) can be calculated and will lead to the relative loop volume change using equation (2).

\[
\Delta V_v = -\Delta V_L
\]

\[
\frac{\Delta V_L}{V_L} = \frac{m_L}{V_L} \left[ \left( \frac{\partial \nu}{\partial T} \right)_p \cdot \Delta T_L + \left( \frac{\partial \nu}{\partial p} \right)_T \cdot \Delta p_L \right] = \frac{1}{v_L} \left[ \left( \frac{\partial \nu}{\partial T} \right)_p \cdot \Delta T_L + \left( \frac{\partial \nu}{\partial p} \right)_T \cdot \Delta p_L \right]
\]

**Table 1.** Partial derivates for the average temperature and pressure increase over the loop.

| \(p_1\) | \(T_1\) | \(\nu_1\) | \(p_2\) | \(T_2\) | \(\nu_2\) | \(\left( \frac{\partial \nu}{\partial T} \right)_p\) | \(\left( \frac{\partial \nu}{\partial p} \right)_T\) |
|--------|--------|---------|--------|--------|---------|-----------------|-----------------|
| MPa    | K      | m³/kg   | MPa    | K      | m³/kg   | m³/kg-K         | m³/kg-bar       |
| 1.32   | 17.2   | 0.01328 | 1.32   | 18.4   | 0.01347 | 0.0001617       |                 |
| 1.32   | 18.4   | 0.01347 | 1.57   | 18.4   | 0.01342 | 0.0001840       |                 |

The 40 K vapor volume change may be calculated according to equation (3). Finally, the necessary initial vapor volume can be evaluated using equation (4) with the derivative of specific volume over pressure for constant entropy (see table 2).

\[
\Delta V_v = m_v \cdot \left( \frac{\partial \nu}{\partial p} \right)_s \cdot \Delta p_L = \frac{V_L}{v_v} \cdot \left( \frac{\partial \nu}{\partial p} \right)_s \cdot \Delta p_L
\]
\[ V_V = \frac{-\Delta V_L \cdot v_V}{\left( \frac{\partial v}{\partial p} \right)_s \cdot \Delta p_L} \]  

(4)

**Table 2.** Isentropic compression of the 40 K vapor fraction.

| State | P  | T   | S    | v    | \( \frac{\partial v}{\partial p} \)_s |
|-------|----|-----|------|------|-----------------------------|
| OFF   | 1.32| 40.00| 16.60| 0.0967| -0.033801 |
| ON    | 1.57| 42.82| 16.60| 0.0883|                 |

Figure 7 shows characteristic data for varying neutron beam powers, heat loads or average temperatures. A linear temperature increase from 17.25 to 18.75 K is being caused by altering the neutron beam power from zero to its maximum value. The loop volume variation is for zero beam power negative and increases with beam power up to ca. 1.5 %. A negative loop volume variation can be interpreted as hypothetical expansion of the 40 K vapor fraction and outflow of cold liquid from the bottom to increase the pressure from 1.32 to 1.57 MPa. In real operation at lower beam powers a higher initial pressure would be chosen or heat would be applied to expand the 40 K vapor to reach 1.57 MPa. Absolute values of initial vapor volume (beam OFF) and respective volume after reaching operating pressure (beam ON) are shown at the top of figure 7. In figure 8 the progressively increasing initial 40 K vapor volume requirement is shown if a smaller isentropic pressure buildup or difference to the operating pressure is favored.

**Figure 7.** Change of vapor volume and mean loop temperature with varying neutron beam power.

**Figure 8.** Necessary initial vapor volume as function of acceptable isentropic pressure increase for full neutron heat load.

### 2.3.2. Actively controlled cold storage vessel.

Instead of only compressing the 40 K vapor fraction in the case of an incoming fluid flow at the bottom of a passive buffer an actively controlled valve could release the same volumetric flow rate of 40 K vapor at the top. This small warm mass flow will be guided through a heat exchanger being cooled by the helium refrigerator and directed to the suction side of the pump (see figure 9). This results in almost no pressure increase compared to the passive storage vessel. If the beam power decreases the cold fluid from the bottom of the vessel has to be guided back to the loop by reestablishing the required vapor volume. A pipe connecting the bottom and top of the vessel could act as a thermosiphon with a small electrical heater.
Figure 9. Actively controlled cold storage vessel: Transition from beam OFF (left) to beam ON (middle) and open control valve for vapor release and cool-down to pump suction side; transition from beam ON (middle) to beam OFF (right) and necessary heating to re-establish vapor fraction.

Equation 5 shows the calculation of the change in loop volume for an actively controlled cold storage vessel which is also being referred in figure 9. The change of average specific volume for a mean temperature increase and constant pressure is listed in table 3.

\[
\Delta V_L = \frac{V_L \cdot (\bar{v}_{ON} - \bar{v}_{OFF})}{\bar{v}_{OFF}}
\]  

(5)

| State | \( p_1 \) [MPa] | \( T_1 \) [K] | \( v_1 \) [m³/kg] | \( p_2 \) [MPa] | \( T_2 \) [K] | \( v_2 \) [m³/kg] | \( v_{mean} \) [m³/kg] |
|-------|-----------------|----------------|------------------|-----------------|----------------|------------------|-------------------|
| OFF   | 1.57            | 17             | 0.01320          | 1.57            | 17.5           | 0.01328          | 0.01324           |
| ON    | 1.57            | 17             | 0.01328          | 20.5            | 0.01380        | 0.01354          |

Table 3. Change of the mean specific volume for the actively controlled vessel.

For the values of the exemplary configuration stated in section 1, a cold loop fluid mass flow of 53.2 g/s enters (equation 6) and a 40 K vapor mass flow of 7.9 g/s (equation 7) has to simultaneously leave the vessel over the heat exchanger to the suction side of the pump over a time period of 12 seconds.

\[
\dot{m}_L = \frac{V_L^2 \cdot (\bar{v}_{ON} - \bar{v}_{OFF})}{\bar{v}_{OFF} \cdot 2V_{pump}}
\]  

(6)

\[
\dot{m}_V = \frac{\Delta V_L \cdot v_v \cdot V_L}{2V_{pump}}
\]  

(7)

By using an actively controlled cold storage vessel, the size of the buffer could be decreased and at the same time significantly reduce the pressure variation compared to that in the passive storage vessel. The large 40 K volume is not necessary to perform a compression because vapor is being released to the main loop. One third to half the size of the passive cold buffer vessel would be sufficient for the actively controlled storage.

3. Transient numerical CFD simulation of the passive storage vessel

Since the ideal process of isentropic compression used in section 2.3.1 of only the 40 K vapor fraction (in the case of the passive vessel) does not take into account a couple of real effects, a transient numerical CFD simulation has been set up. Figure 10 shows the geometry and thermodynamic state related to the isentropic compression process in figure 6.
The most significant effects which have been previously neglected may be the compressible behavior of the transition zone from 17 to 40 K, vertical heat conduction and forced mixing of areas of different temperature due to the incoming fluid flow. Heat conduction along and heat transfer over the wall of the vessel has not been implemented so far. Real gas fluid properties have been implemented to allow numerical simulations between temperatures from 16 to 100 K and pressures from 1.3 to 2.0 MPa. All fluid properties have been implemented using high resolution tables as function of pressure and temperature. Density data has been fitted with a slightly decreased slope close to the critical temperature of 33 K. The relative error is around ±10 % for the temperature range around 32 to 36 K. For higher and lower temperatures the relative error decreases very quickly below ±1 %. Figure 10 shows the absolute values for the equation of state and the customized fit at 1.3 MPa. The isobaric heat capacity has been clipped for numerical reasons at 100000 kJ/kg·K which results in a relative error of -6 % for the enthalpy difference from 16 to 40 K at 1.5 MPa. Viscosity and thermal conductivity have been implemented without any changes. Figure 11 shows an inflow and compression process of the passive vessel for full power beam introduction in the main loop and related increase of the mean loop temperature.

For 12 seconds a mass flow of 30 g/s at 17.25 K enters the vessel. The pressure increase of 0.08 MPa is less compared to the ideal isentropic compression with 0.25 MPa in section 2.2. For the ideal calculation the compressible feature of the transition zone from 17.25 to 40 K was neglected.
Furthermore, the incoming fluid forms a small plug (visible in the temperature distribution of figure 11 at six seconds) which mixes the lower fraction of the transition zone and leads to a mitigating effect on the pressure buildup. Vertical heat conduction would have the same effect. The following outflow process and expansion ends at a slightly smaller pressure than the initial value. The compressible feature of the transition zone has been identified as the most significant influence for the difference between ideal process in section 2.2.1 and the numerical CFD simulation.

4. Conclusion
Different methods to cope with heat load or mean temperature changes in a closed liquid hydrogen circuit and the resulting pressure fluctuations have been presented. The concept of cold storage vessel in form of a passive or an actively controlled buffer has been discussed in detail. A reasonable sized cold vertical buffer including a temperature gradient from the bottom to the top can take or release mass in case of a forced change in the main loop’s specific volume. The actively controlled storage vessel would outperform the passive one by enabling a smaller pressure variation and less required overall volume. The reason for that is the release, cool-down and introduction of vapor in the main loop instead of requiring a large vapor volume for a small pressure increase due to compression. A numerical CFD simulation has been set up to understand the hydrodynamic behavior of a passive cold storage vessel in more detail. It could be shown that the temperature gradient will be maintained during the inflow process and that the pressure increase is less compared to the ideal case of isentropic compression of the higher temperature vapor at the top of the buffer.

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References
[1] Crabtree J A 2005 Proc. of ICANS XVII Passive Pressure Control for the SNS Hydrogen Moderator System pp 306-14
[2] Tatsumoto H, Aso T, Ohtsu K 2009 Proc. of CEC Pressure fluctuation behavior in the cryogenic hydrogen system caused by a 100 kW proton beam injection pp 289-96
[3] Lemmon E W, Huber M L, McLinden M O 2013 NIST Standard Reference Database 23 Reference Fluid Thermodynamic and Transport Properties-REFPROP Version 9.1 National Institute of Standards and Technology Standard Reference Data Program Gaithersburg
[4] Leachman J W, Jacobsen R T, Penoncello S G, Lemmon E W 2009 J. Phys. Chem. Ref. Data 38 (3) Fundamental Equations of State for Parahydrogen, Normal Hydrogen, and Orthohydrogen pp 721-48
[5] Quack H, Dhard C P 2012 Proc. of ICEC 24-ICMC Storage of Supercritical Helium pp 295-8