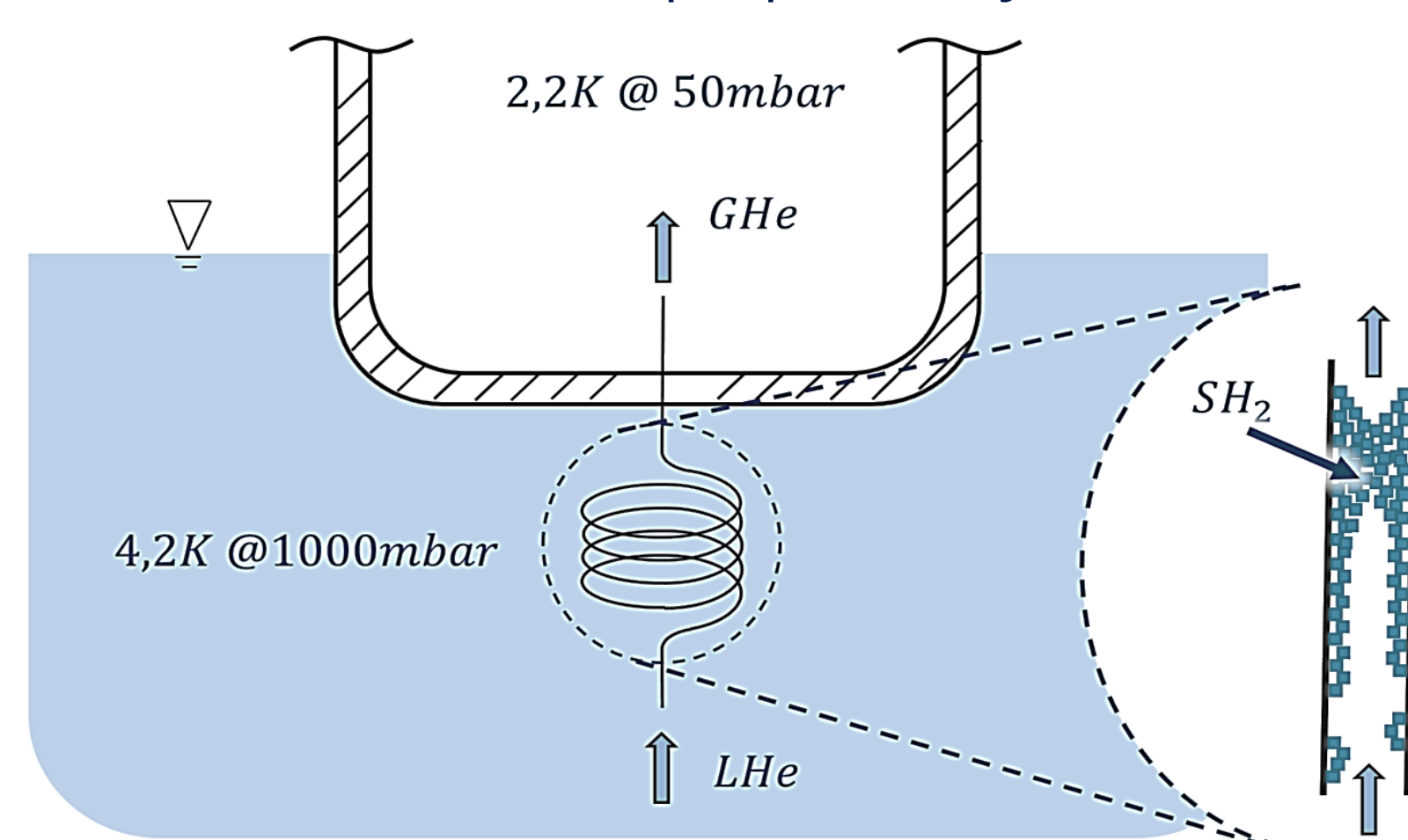


Motivation

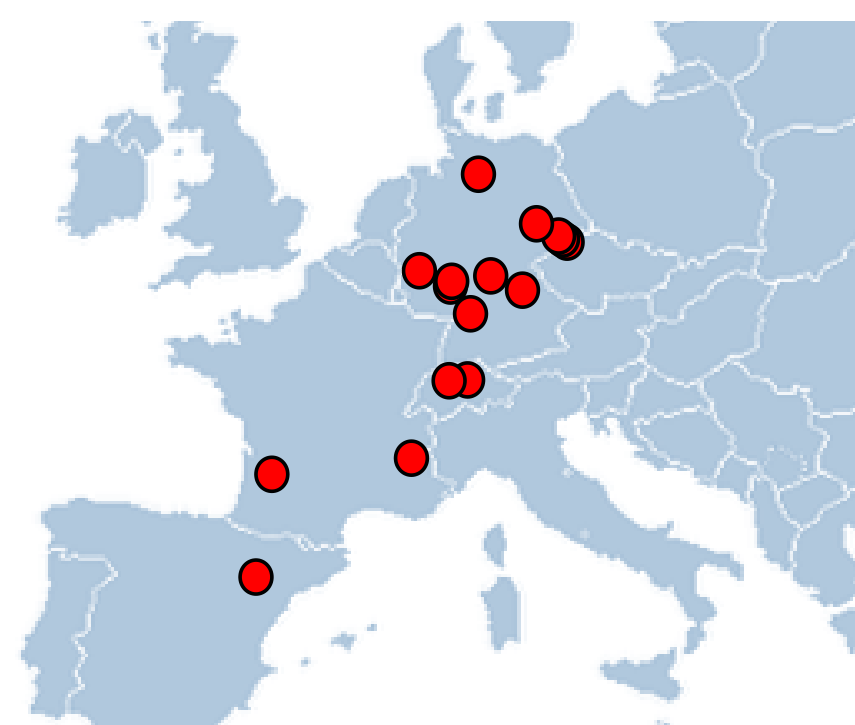
Liquid helium is used in cryogenic laboratories worldwide. In recent years there has been an increase in equipment failures due to hydrogen blockages.

To achieve temperatures below the saturation temperature of 4.2 K, the pressure of the helium will be reduced. For this purpose, cryostat internal narrow throttling passages (capillaries) or throttling valves are used. Blockage problems have been occurring for several years now. Hydrogen precipitates in solid form during the helium evaporation and accumulates at the throttle location.



Hydrogen separates from the two phase helium flow and accumulates as a solid phase on the inner walls. This creates a solid, clogging block.

- Many problem reports from Germany and Europe.
- First reports came up in 2005.
- Blockages arising typically within a few hours of operation.
- Reports also exists from Asia.

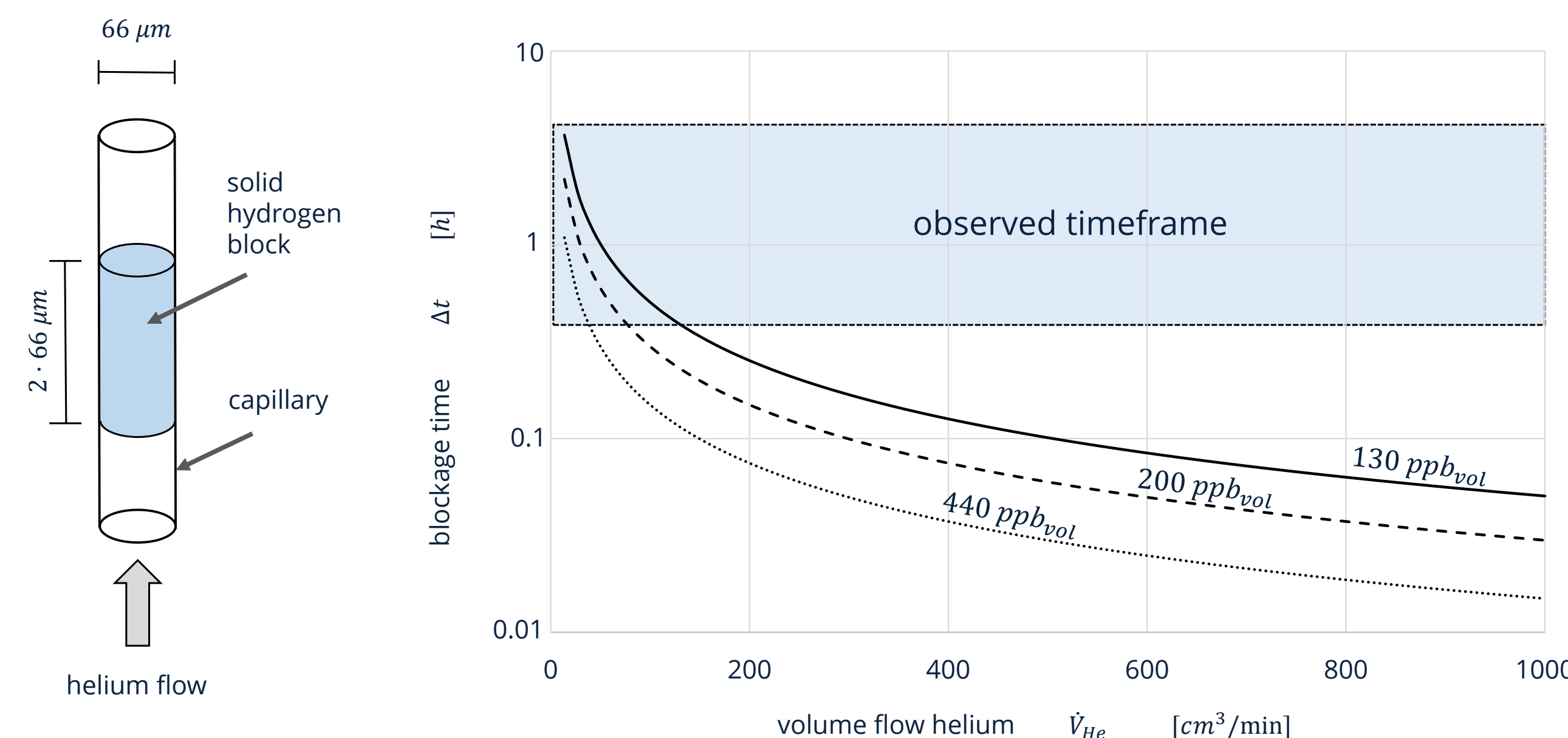


Plausibility of hydrogen as cause

There are various indications that hydrogen is the reason for the blockage problem:

- A blockage can be eliminated by warming up the affected location to 20 ... 30 K (triple point of hydrogen: 14 K)
- An increased amount of hydrogen was measured spectrometrically at the moment of dissolution of a blockade.

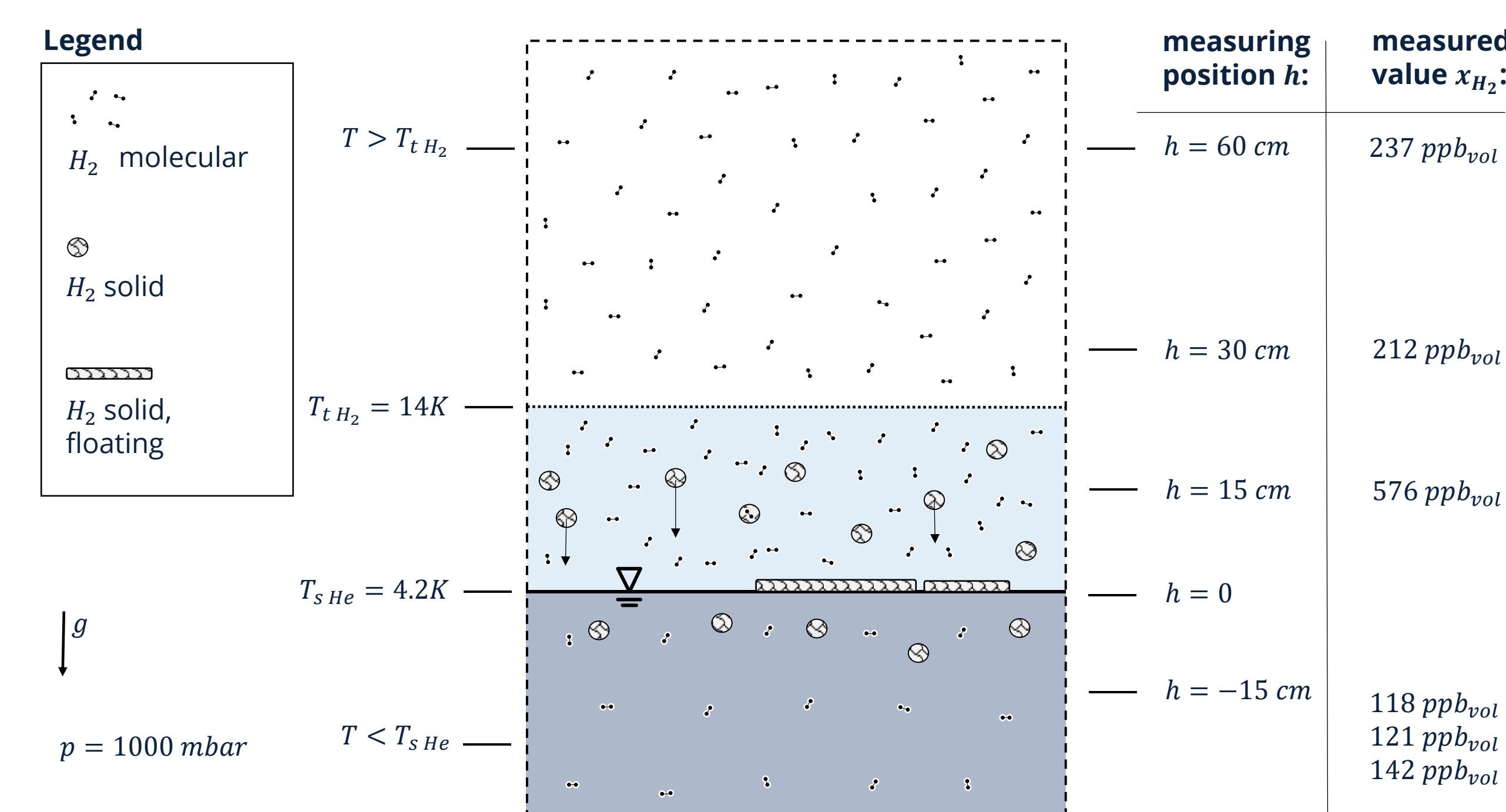
A simple calculation demonstrates that even small quantities of hydrogen can be sufficient to cause a blockage:



The left scheme shows a simple model of the blockade build up. In the capillary through which the helium flow passes, all hydrogen contamination will freeze out and form a cylindrical solid block. The diagram gives the results based on this simple calculation model. The curves show the duration of the blockade build up, based on different measurement values. Even if not all of the hydrogen participates in the blockade, the contents are more than sufficient.

System Helium/Hydrogen

For understanding the helium/hydrogen phenomena a macroscopic visualization model was developed:



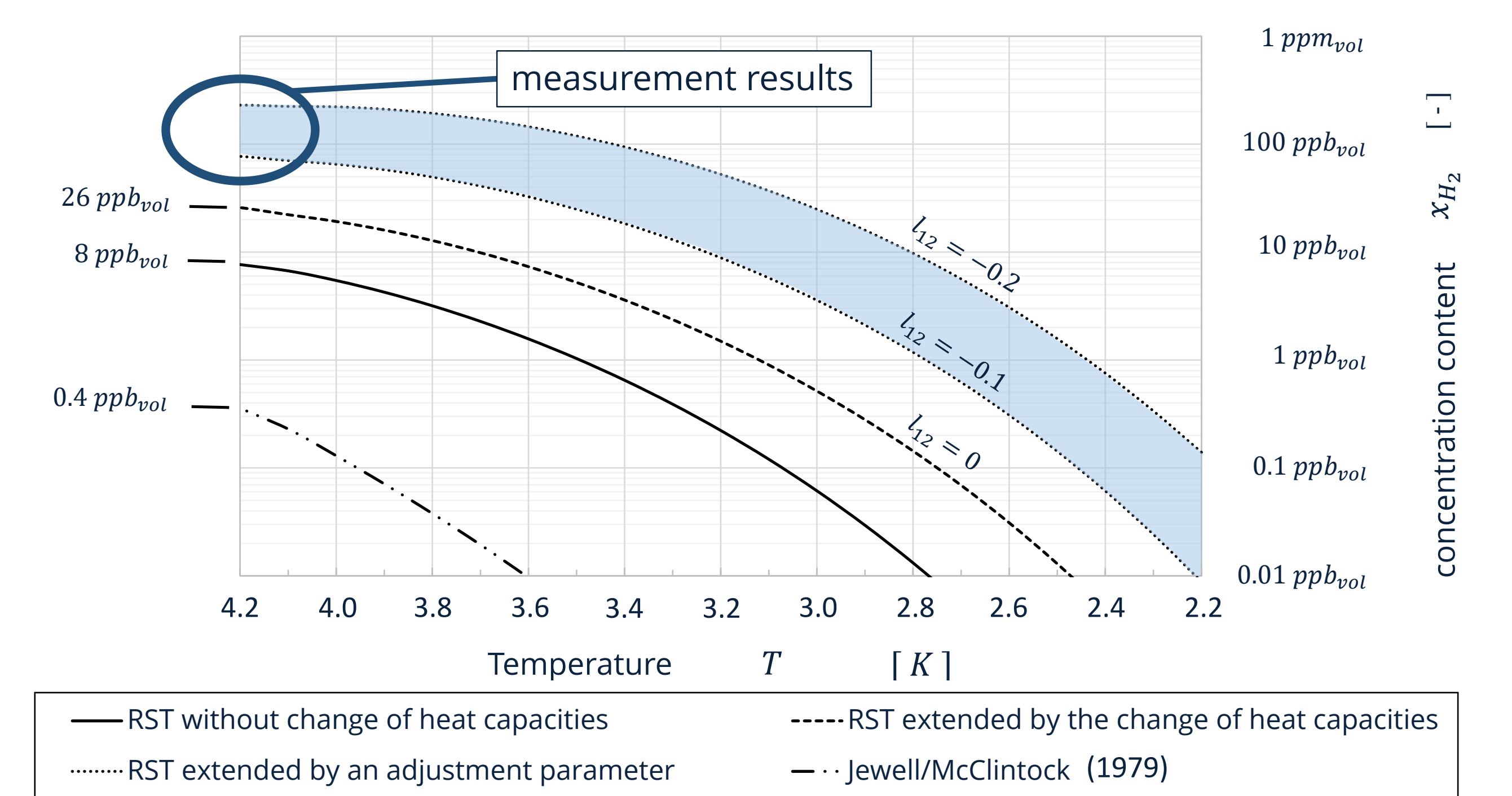
According to the model, the reservoir is divided in different temperature ranges in which the hydrogen behaves differently. The liquid helium contains both dissolved and crystalline hydrogen. Measurements were carried out at different positions within a helium dewar. Gas chromatography was used for this purpose. These values provide the range of contamination quantities.

Probably, dissolved and crystalline hydrogen can coexist in liquid helium and are extremely hard to distinguish. A calculation of the solubility limit is a first step in estimating the proportions. The Regular Solution Theory (RST) is used:

$$\ln x_i^L = \underbrace{-\frac{\Delta H_{f,m,i}(T_{f,i})}{RT}}_{\text{ideal solubility}} \left(1 - \frac{T}{T_{f,i}}\right) + \underbrace{-\frac{\Delta c_{p,m,i}^{S \rightarrow L}}{R} \left(1 - \frac{T_{f,i}}{T}\right) + \frac{\Delta c_{p,m,i}^{S \rightarrow L}}{R} \ln \frac{T}{T_{f,i}}}_{\text{change of heat capacities}} - \ln \gamma_i^L$$

$$RT \cdot \ln \gamma_i^L = V_{m,i}^L \phi_j^2 [(\delta_i - \delta_j)^2 + 2l_{ij} \delta_i \delta_j]$$

activity coefficient activity coefficient binary adjustment parameter



The calculation results in 26 ppb at 4.2 K. In the literature can be found the application of the RST by Jewell and McClintock from 1979, which is presented for comparison. The binary adjustment parameter l_{12} must be set in the range from -0.1 to -0.2. Experience has shown that such values for l_{12} are one order of magnitude too high. This could be an indication that beside the dissolved hydrogen, crystalline hydrogen can exist in liquid helium.