

Capacitive density measurement for supercritical hydrogen

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Abstract. A new approach for automotive hydrogen storage systems is the so-called cryo-compressed hydrogen storage (CCH₂). It has a potential for increased energy densities and thus bigger hydrogen amounts onboard, which is the main attractiveness for car manufacturers such as BMW. This system has further advantages in terms of safety, refueling and cooling potential.

The current filling level measurement by means of pressure and temperature measurement and subsequent density calculation faces challenges especially in terms of precision. A promising alternative is the capacitive gauge. This measuring principle can determine the filling level of the CCH₂ tank with significantly smaller tolerances. The measuring principle is based on different dielectric constants of gaseous and liquid hydrogen. These differences are successfully leveraged in liquid hydrogen storage systems (LH₂).

The present theoretical analysis shows that the dielectric values of CCH₂ in the relevant operating range are comparable to LH₂, thus achieving similarly good accuracy. The present work discusses embodiments and implementations for such a sensor in the CCH₂ tank.

1. Introduction

A new approach for hydrogen storage systems for vehicle applications is the so-called Cryo-compressed hydrogen tank (CCH₂ storage vessel). With these, hydrogen is stored at a pressure between ca. 20 bar and 300 bar at temperatures between ca. 33 K and elevated temperatures up to ambient conditions. Thus the hydrogen is always in the state of a one-phase supercritical fluid. Especially at low temperatures (after refueling and for frequent drivers) fairly high storage densities of up to 72 g/l can be realized. Additionally, even in case of long parking periods, an appreciable amount of high-pressure hydrogen gas is left to ensure mobility. Thus, advantages both of the low-pressure LH₂ two-phase tanks and of warm high pressure hydrogen tanks are combined.

An essential feature of a storage system is the level measurement. New solutions are needed for appropriate and precise hydrogen content metering in such CCH₂ tanks. Superconducting level gauges developed for LH₂ tanks can't be applied because these essentially detect the liquid/gas phase boundary, which does not exist in CCH₂ storage systems. Furthermore simple density deductions from combined pressure and temperature measurement struggle due to possible pronounced thermal stratification of the stored hydrogen.

The present theoretical analysis investigates capacitive gauges as a viable alternative. These were developed originally as level gauges for low-pressure two-phase hydrogen tanks as shown in figure 1. The measuring principle is based on different values of the dielectric constant for liquid and gaseous



hydrogen. An application in a CcH2 system uses the dependency between fluid density and dielectric constant. The obtainable accuracy in this case is investigated.

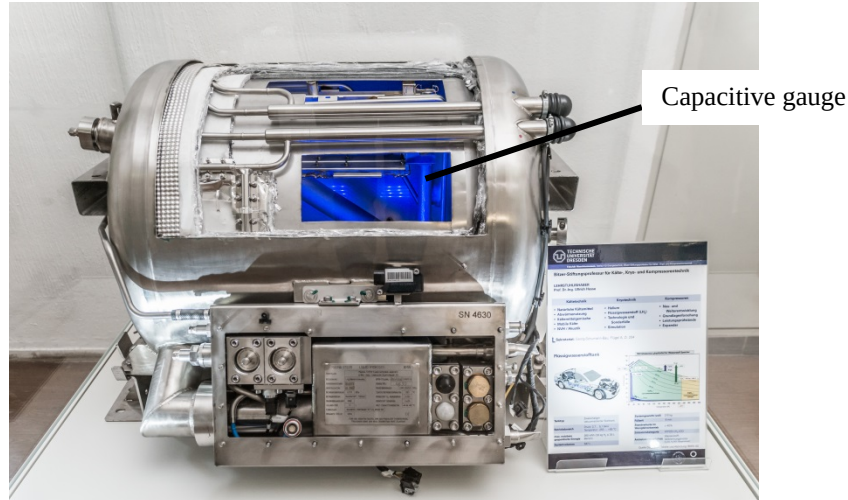


Figure 1. LH₂ storage vessel for automotive application with a capacitive level gauge. Hydrogen capacity approx. 8 kg.

2. Investigated configuration and fluid data

The investigated capacitive level gauge, consists of 3 coaxial tubes of 570 mm length. It is designed to detect the liquid level inside the two-phase storage vessel. Therefore, it is placed vertically inside the vessel.

A capacitor partially immersed into liquid corresponds to two capacitors in parallel arrangement; one with gaseous hydrogen and the other one with liquid hydrogen as the dielectric medium (see figure 2). Thus, its total capacity is the sum of the sub-capacitor in the gaseous phase and in the liquid phase.

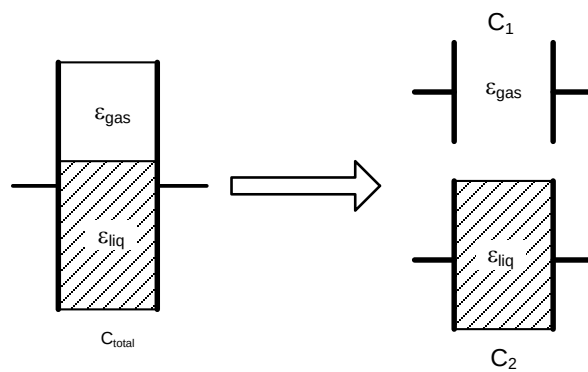


Figure 2. Equivalent circuit of a partially immersed capacitor in a liquid and gaseous phase.

The capacity of a cylindrical capacitor is given by:

$$C_{total} = 2\pi\epsilon_0\epsilon_r \left(\frac{l}{\ln\left(\frac{R_2}{R_1}\right)} \right) \quad (1)$$

With ϵ_0 being the dielectric constant ($\epsilon_0 \approx 8.854 \times 10^{-12} \text{ Fm}^{-1}$), ϵ_r the relative dielectric constant, l is the length of the capacitor, R_2 the outer and R_1 the inner diameter of the cylindrical probe.

This equation applied to the present model of two concentrically arranged, cylindrical capacitors, is shown in equation (2). Here, h describes the height of the liquid, respectively the liquid hydrogen level, l the length of the capacitor, $\epsilon_{r, \text{gas}}$ the relative dielectric constant for gaseous hydrogen, and $\epsilon_{r, \text{liq}}$ the relative dielectric constant for liquid hydrogen. The geometrical dimensions R_1 , R_2 , R_3 and R_4 are stated in table 1.

$$C_{\text{total}} = 2\pi\epsilon_0 \cdot (\epsilon_{r, \text{gas}}(l - h) + \epsilon_{r, \text{liq}}h) \cdot \left(\frac{1}{\ln\left(\frac{R_2}{R_1}\right)} + \frac{1}{\ln\left(\frac{R_4}{R_3}\right)} \right) \quad (2)$$

For supercritical hydrogen equation (2) simplifies due to the dissolved phase boundary to

$$C_{\text{total}} = 2\pi\epsilon_0\epsilon_{r, \text{gas}} \cdot l \cdot \left(\frac{1}{\ln\left(\frac{R_2}{R_1}\right)} + \frac{1}{\ln\left(\frac{R_4}{R_3}\right)} \right) \quad (3)$$

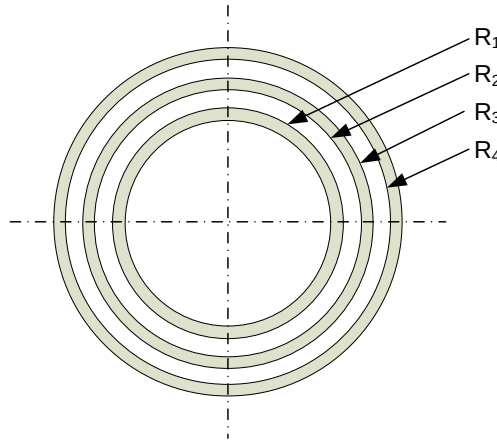


Figure 3. Relevant dimensions of the capacitor (consisting of three concentric tubes) to calculate its capacity according to equation (3).

Table 1. Dimensions of the investigated capacitive level gauge (shown in figure 3).

dimension	value [mm]
R_1	9
R_2	11.5
R_3	12.5
R_4	14
length	570

2.1. Fluid data

The hydrogen molecule can exist in two different spin configurations: orthohydrogen (o-H₂) and parahydrogen (p-H₂). Both configurations coexist with a ratio of ortho- to parahydrogen depending on the temperature. An equilibrium mixture of 25 % p-H₂ and 75 % o-H₂ is called normal-hydrogen (n-H₂). According to Pashkov et al. [2] the relative dielectric constants for o-H₂ and p-H₂ are very similar. Thus, for this investigation the fluid data for p-H₂ are used. In table 2 relevant data for liquid and gaseous hydrogen in equilibrium state is listed.

Table 2. Relative permittivity for parahydrogen in liquid and gaseous phase at equilibrium state over a pressure range from 1 bar to 6 bar [3].

p [bar]	T _{boil} [K]	ε _{r, gas}	ε _{r, liquid}
1	20.2	1.0040	1.2306
2	22.8	1.0075	1.2193
3	24.6	1.0111	1.2105
4	26.0	1.0148	1.2027
5	27.1	1.0186	1.1954
6	28.1	1.0227	1.1813

According to Steward [1] equation (3) is given for the dielectric constant at a given density. This correlation is valid in a density range of 0 g/l to 80 g/l. The parameter P is fitted by a quadratic function shown in equation (4). Figure 4 shows a diagram of the relative dielectric constant over the hydrogen density.

$$\varepsilon = \frac{(1+2 \cdot P \cdot \rho)}{(1-P \cdot \rho)} \quad (4)$$

$$P^{-1} = A + B \cdot \rho + C \cdot \rho^2 \quad (5)$$

With: A = 0.99575 g/cm³
 B = -0.09069
 C = 1.1227 cm³/g

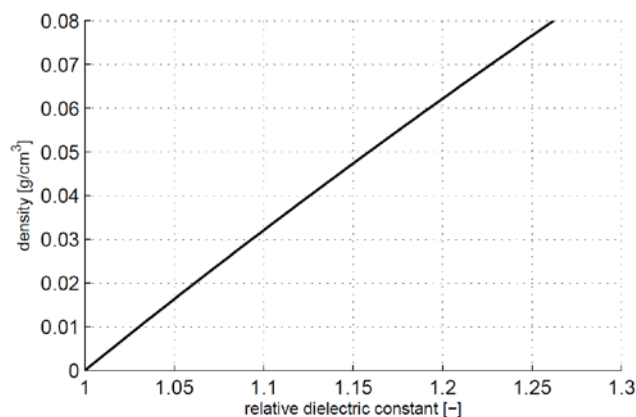


Figure 4. Density over the relative dielectric constant for parahydrogen [2]

3. Analysis

In table 3 the maximum and minimum amplitude of the capacitive level sensor are given. For the LH₂ system, depending on the pressure level, the signal difference from maximum to minimum level is 42.6 pF to 62.2 pF. This is a direct measure of the height of the liquid level. The stored amount of hydrogen is derived from that by geometry factors. Figure 5 shows the resolution in [g pF⁻¹] for a cylindrically shaped storage vessel (ø 570 mm, length 500 mm). The storage tank is oriented horizontally and the sensor is installed perfectly vertical direction. Corresponding to the circular cross section, the resolution is symmetric with a maximum in the centre, where one mm liquid represents the biggest hydrogen volume. The resolution is between 197.3 g pF⁻¹ and 36 g pF⁻¹.

The range of the measured capacity for a CcH₂ storage vessel is 75.2 pF from empty to full. For the described CcH₂ storage system at the maximum filling level a density of 84.7 g l⁻¹ is reached and 1.6 g l⁻¹ at minimum filling level (i.e. empty tank). The dielectric values in the operating range between 30 K/300 bar and 300 K/23 bar are 1.0056 and 1.2790 respectively. This corresponds to a resolution of 0.91 g l⁻¹ pF⁻¹. Applied to the same volume of approx. 128 l a resolution of the stored hydrogen mass of 116.1 g pF⁻¹ is achieved.

Table 3. Amplitude of a capacitive level gauge in a LH₂ and CcH₂ system.

	p [bar]	max. level [pF]	min. level [pF]	(max – min) [pF]	Resolution [mm pF ⁻¹]
LH ₂	1	338.3	276.2	62.2	9.1
	2	335.5	277.1	58.4	9.8
	3	333.0	278.1	54.9	10.4
	4	330.9	279.1	51.8	11.0
	5	328.9	280.1	48.8	11.7
	6	327.0	281.2	45.8	12.5
	7	325.1	282.5	42.6	13.4
CcH ₂	30 ... 300	351.8 (30 K)	276.6 (300 K)	75.2	1.1 g l ⁻¹ pF ⁻¹

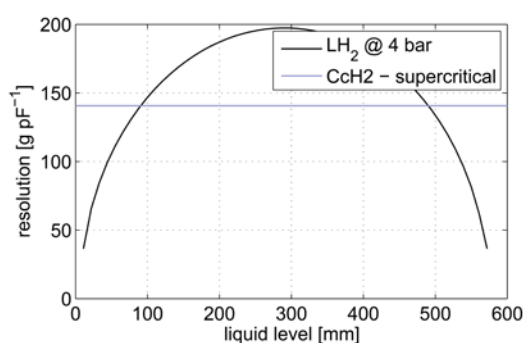


Figure 5. Resolution of a capacitive level gauge inside a LH₂ dewar. The geometry of the latter one is simplified to a cylindrical shape (D = 570 mm, l = 500 mm) And the sensor is installed vertically.

4. Conclusion

A capacitive gauge designed for liquid hydrogen tanks was theoretically analyzed for a CcH₂ storage vessel. The correlation between density and dielectric constant leads to a resolution of 140.8 g pF⁻¹. This value scales with the length of the probe. A two times longer probe could achieve a resolution of 58.05 g pF⁻¹. This effect is even more prominent for the fluid filled gap between the respective

cylinders. If e.g. reduced from 1.5 mm to 1 mm each, the resolution is enhanced drastically to 91.4 g pF^{-1} .

Since the dielectric constant correlates directly to the fluid density, it is independent from geometrical arrangement inside the vessel.

CcH₂ storage vessels show a pronounced thermal stratification, but a properly arranged capacitive probe along all density layers naturally determines the average fluid density along the length of the probe.

This investigation indicates that the capacitive gauge could be a viable alternative to existing combination of pressure and temperature measurement for precise hydrogen content metering.

5. References

- [1] Stewart J W 1964 Dielectric polarizability of fluid para-hydrogen *J. Chem. Phys.* **40** 3291-3306
- [2] Pashkov V V and Lobko M P 1980 Dielectric properties of liquid hydrogen *Advances in Cryogenic Engineering* **35 A** 709-714
- [3] Lemmon E W, Huber M L and McLinden M O 2013 NIST reference database 23: Reference fluid thermodynamic and transport properties REFPROP 9.1 *National Institute of Standards and Technology*, Gaithersburg