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LONG-TERM HYDROGEN STORAGE UTILIZING CFC GAS-STORAGE SYSTEM

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ABSTRACT

When molecules or atoms are attached to a surface, they can do so using one of two main methods: chemisorption or physisorption. This project focuses on utilizing the novel technology of the Cryogenic Flux Capacitor (CFC) assembly to store hydrogen through the process of physical adsorption. The thermally controlled vessel will have varying concentrations of orthohydrogen and parahydrogen during adsorption and desorption of the hydrogen molecules from the surface. Charging of the molecules will occur at around 19 K while discharging them is set to be at 77 K. Due to parahydrogen's lower level of energy, it's more desired as it will allow for the maximum amount of energy to be stored. On the other hand, orthohydrogen is preferential during physisorption at low temperatures. Thus, we will need to find an equilibrium mixture of hydrogen where we get enough hydrogen atoms physiosorbed to store energy while still minimizing hydrogen in the ortho state as it would use an unnecessary amount of energy which could've been stored inside the vessel long-term.

Keywords: Aerogels, cryogels, orthohydrogen, parahydrogen, normal hydrogen, physisorption, adsorption

NOMENCLATURE

CFC Cryogenic Flux Capacitor
ASME American Society of Mechanical Engineering

c_p Heat Capacity (Constant Pressure)

c_v Heat Capacity (Constant Volume)

μ Dynamic Viscosity
ρ Density
k Thermal conductivity
Pr Prandtl Number
s Entropy
h Enthalpy

1. INTRODUCTION
1.1 Hydrogen Storage

With the increased demand for energy, scientists and companies are looking for alternate methods to generate and store energy in the most economically efficient ways. Subsequently, hydrogen has been of interest for many for its economic efficiency and long-term capability of storing energy.

It can be pressurized as a gas or stored at low-pressure conditions in the liquid form. The vessels used to store hydrogen have to be designed following the ASME code for boilers and pressure vessels. For that reason and for its cutting-edge technology, we decided to use the CFC model designed by NASA where they use the adsorption process to charge (adsorption) and discharge (desorption) hydrogen molecules inside the vessel.

1.2 Physisorption

When molecules or atoms are attached to a surface, they can do so using one of two main methods: chemisorption or physisorption –a contraction of chemical adsorption and physical adsorption, respectively– [2]. These methods are possibly the most promising hydrogen storage bonding mechanisms as they allow for storage of large amounts of hydrogen in relatively small volumes [28]. The molecules will form chemical bonds with the surface of the substrate in a chemisorption process [2] similar to a bond created between atoms in a molecule [29]. The hydrogen molecules break into hydrogen atoms before integrating into the material of the surface during this process [28]. If the interactions between the chemisorbed molecules and the surface atoms are strong enough, these bonds can be broken [2].

On the other hand, in physisorption, hydrogen atoms or molecules get attached to the surface, diminishing the distance between the molecules and the surface. Subsequently, allowing to control and manage thermal issues during the discharge and discharge process [28]. The two materials involved in the physisorption (i.e. adsorption) are the substance whose molecules get adsorbed and the one whose surface hosts the adsorption process. They are called the adsorbate and adsorbent/substrate, respectively. Interactions between the two are caused by van der Waals forces [2]. Such weak interactions have a long range. In a process referred to as accommodation, the small amount of energy involved in physisorption gets dispersed into vibrations [2].

Adsorption is an exothermic process [29]. When knowing the heat capacity of a sample, the rise of temperature in that sample can be an indicator and a measuring tool for the enthalpy of physisorption. A physiosorbed molecule retains its properties as the change in enthalpy during the process is very small and not enough to break any bonds. However, it might experience some distortion caused by the surface where the process is occurring. Chemisorption, on the other hand, has

much greater enthalpy measurements [2]. The adsorbent material should be light-weight and have sufficient bonding sites [28].

1.3 Ortho- and Para-Hydrogen

Taking into consideration the change of temperature that will occur in the vessel, it was found that we also need to consider multiple states of hydrogen: orthohydrogen and parahydrogen.

To acquire the quantum mechanical property nuclear spin, the nucleus has to have an odd number of protons/neutrons. Since there's only one proton in the nucleus of a hydrogen atom, it possesses nuclear spin. Note that, although we are considering protons, this property does not refer to the actual rotation of the proton but rather it's just a useful analogy to explain this property [9]. Basing off that analogy, just like electrons spin around the nucleus of the atom, protons also spin inside the nucleus. The direction in which the protons are spinning is the direction in which the nucleus and, therefore also, the atom is spinning. A molecule of dihydrogen contains two atoms of hydrogen. Based on the direction of nuclear spin of each nucleus in a molecule, we can categorize hydrogen molecules into orthohydrogen molecules and parahydrogen molecules. The prefix "ortho" is used to describe the case where both nuclei are spinning in the same direction. Parahydrogen, on the other hand, has two atoms of hydrogen that are spinning in opposite directions [10].

Ordinary hydrogen is an equilibrium mixture of both orthohydrogen and parahydrogen. At low temperatures, parahydrogen is the form prevalent, but orthohydrogen has a dominant concentration at room temperature and high temperatures overall [5][6][10].

While there's a difference in potential parameters between orthohydrogen and parahydrogen, this difference is not determined in most modern statistical thermodynamics' texts due to its insignificance [10]. In our case, we will be looking into parahydrogen as the pressure in that state tends to fluctuate slightly more [15].

2. MATERIALS AND METHODS

For orthohydrogen, the nuclear wavefunction is symmetric, while for parahydrogen it's antisymmetric. With this in mind, note that the magnetic quantum number of protons is $\frac{1}{2}$. Subsequently, and using quantum mechanical addition, for antisymmetric/antiparallel spins the net spin will be 0, whereas the net spin of symmetrical/parallel spins is 1 [2][5][10][11].

Orthohydrogen has an odd rotational quantum number while parahydrogen has an even number [2][6][8]. To describe the number of possible substates for the nuclear wavefunction for para- and ortho- hydrogen, we use "singlet" and "triplet" spin multiplicities. Due to the asymmetry of parahydrogen, it only has one possible substate and thus it's called a singlet. In contrast, the symmetry of orthohydrogen causes it to have three possible substates and is therefore called a triplet and is also higher in as a result of that [2][9][10][11]. For spin multiplicities, the multiplicity rule (# of unpaired electrons + 1)

is applied to identify them [2][5][9]. For the net spin = S and using the multiplicity rule for orthohydrogen: 2S + 1 = 2*1 + 1 = 3 (i.e. triplet with 2 unpaired electrons), on the other hand, if we apply the same rule for parahydrogen then we will have 2S + 0 = 2*0 + 1 = 1 (i.e. singlet with 0 unpaired electrons).

At 19 K, equilibrium hydrogen doesn't have sufficient thermal energy to populate a significant amount of higher energy states (triplets) which results in 99.75% parahydrogen since it has a lower energy level (singlets) [2][7][10][16]. Orthohydrogen percentage is ~1% upon cooling the temperature to 20 K (liquid hydrogen) [3]. If the temperature increased to 80 K, for instance, the equilibrium concentration ratio will be approximately 1:1 [10]. As the temperature approaches room temperature (around 293 K), the thermal energy the hydrogen possesses is enough to get a mixture concentration of 75% orthohydrogen and 25% parahydrogen.

This 3:1 ratio is maintained in temperatures higher than room temperature and is often referred to as normal hydrogen [2][5][7][10][16]. In essence, parahydrogen and normal hydrogen have similar properties [15]. However, note that it's not possible to get more than 75% concentration of orthohydrogen and it cannot be in pure form without special catalysts or distillation techniques [10][16].

Temperature, K	Parahydrogen Percentage, %
10	99.9999
20	99.821
30	97.021
40	88.727
50	77.054
60	65.569
70	55.991
80	48.537
90	42.882
100	38.620
120	32.959
150	28.603
200	25.974
250	25.264
300	25.072

TABLE 1: Ortho-para hydrogen composition at equilibrium. As reported in [19] Hust et al. (1965).
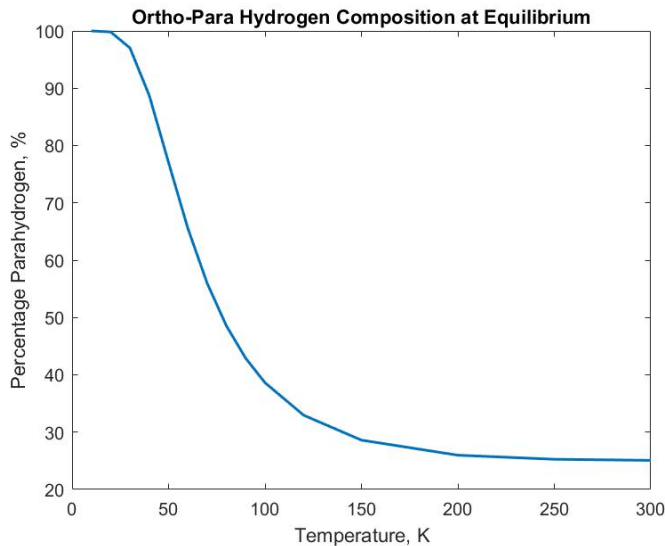
FIGURE 1: Curve of ortho-para hydrogen composition at equilibrium through a range of temperatures.

In the case of normal hydrogen liquefication, orthohydrogen converts abruptly to parahydrogen through an exothermic reaction, which takes the role of a heat source [15][17]. When considering long-term storage, and depending on the ambient temperature, the boil off rate tends to be significant due to the heat produced by the "heat source" [15]. When the temperature is at 77 K, the heat of conversion is 519 kJ/kg, but at any other temperature lower than 77 K it is 523 kJ/kg. Keep in mind that the latent heat of vaporization of normal hydrogen is 451.9 kJ/kg at the normal boiling point, which means both of the heat of conversion values are greater than that heat of vaporization value [17].

The temperature at the maximum point of hydrogen density/power consumption curve is marginally higher when considering ortho-to-para hydrogen conversion compared to when it's not considered [16]. If we make the assumption that no conversion between the para- and ortho-phases of hydrogen occurs, then we would be making a conservative assumption as the worst-case scenario for the vessel is the absence of the minimizer of the heating in the vessel due to heat leakage [13].

However, if we desire to avoid conversion during the adsorption process, then we may equilibrate the hydrogen at the same temperature prior to inserting it in the vessel [14]. On the other hand, when wanting to accelerate ortho-to-para conversion, we would need to insure the existence of unpaired electrons in a great quantity at the edges and the defect points as well as the presence of oxygen atoms in the vessel [3]. The main attribute to ortho-to-para hydrogen conversion can be considered the magnetic dipole interaction of orthohydrogen molecules [12].

We are probably introducing hydrogen at its equilibrium state into the vessel, at near-cryogenic temperatures, meaning the hydrogen mixture will have a high concentration of parahydrogen [13]. As the temperature increases, parahydrogen converts into orthohydrogen, and thus

the parahydrogen concentration decreases. However, the conversion from parahydrogen to orthohydrogen is endothermic, as a result of that the vessel remains cold and the heating of the vessel gets delayed, reducing hydrogen losses from the vessel [1][13].

Furthermore, the para-to-ortho conversion rate is often slow. Subsequently, the equilibration time can take several days [1][5][6][10][13]. It was estimated that the rate of para-to-ortho conversion for normal hydrogen at 20 K (liquid) is ~0.7% per hour [3][5].

When storing hydrogen, it's recommended for the temperature parameters to be between 35-110 K and for the pressure to be within the 5-70 MPa range regardless of ortho-to-para hydrogen conversion [16]. For those parameters, the corresponding densities for hydrogen are 60.0-71.5 kg/m^3.

The value of parahydrogen concentration, at equilibrium, at 300 K is ~25% and ~99.8% at 20 K [12][16]. It can be interpreted from that the existence of a considerable amount of conversion enthalpy, at greatly low temperatures above all. Considering parahydrogen at 20 K, its vaporization enthalpy will be found to be around 447 kJ/kg [16].

When looking into the adsorption process, it was observed in multiple experiments that there is preferentiality of orthohydrogen adsorption as opposed to parahydrogen adsorption. One of the experiments used TiO_2 (rutile) and charcoals as adsorbents while another focused-on studying adsorbed hydrogen molecules on amorphous ice. This later led to the conclusion that orthohydrogen is more strongly adsorbed than parahydrogen when it comes to adsorption on cold solid surfaces [4][14].

It was suggested that the preferential adsorption effect on orthohydrogen mainly connected to the rotation of the adsorbed hydrogen. On cold surfaces, the almost spherical rotational wavefunction of parahydrogen is unable to detect the anisotropic portion of the potential. On the other hand, and due to anisotropy, the non-spherical wavefunction of orthohydrogen possesses additional binding energy when compared to parahydrogen [4][14].

Nevertheless, it wasn't confirmed how much effect other factors- particularly the fact that when parahydrogen is in the lowest rotational state (non-rotating molecule), it has less polarizability than a rotating molecule- can have on difference in adsorption of ortho- and para-hydrogen [14].

Ref. [4] studied adsorbed normal hydrogen in amorphous ice at 12 K over time and concluded that orthohydrogen is expected to have a half-life of about 116 min on average.

Additionally, they showed that after 300 min of deposing hydrogen into the vessel the mixture was composed largely of parahydrogen. It's worth noting that this study was based on a few assumptions: all molecules of H_2 stick to the adsorbent after hitting its surface, the adsorbed hydrogen and the surface were at thermal equilibrium, and the thermal evaporation rate is equal to that of the molecules sticking to the surface. The adsorbed layer of hydrogen tends to maintain the same physical properties (e.g. composition, temperature,

3

evaporation rate and such). It was also reported that ~60% of H_2 molecules could stick in an area of a Cu (100) surface that's already covered with H_2 [4]. In fact, copper, along with a number of other diamagnetic metals, have a surface that accelerates the ortho-to-para conversion process [12].

A pure-parahydrogen model was made in effort to compare its results with the collected data. The number of adsorbate H_2 molecules was 408±4 compared to the previous 454±4 due to the parahydrogen molecules having lower binding energy. Indeed, when considering the orientational distribution of orthohydrogen, orthohydrogen was found to have a large localization in the direction of minimum potential energy while parahydrogen is broadly delocalized [4].

Although the absorption cross section for para- and ortho-hydrogen is the same, the total neutron scattering cross section for orthohydrogen is 80 (10-28 m^2/atom) while it's less than 0.9 (10-28 m^2/atom) for parahydrogen. This means that any significant decrease in neutron scattering intensity can be attributed to ortho-to-para conversion [3].

3. RESULTS AND DISCUSSION

Some mechanical and thermal properties of orthohydrogen, parahydrogen, and normal hydrogen are considered in tables 1 and 2 at room temperature and 77 K, respectively. Remarkable differences can be observed between those different states of hydrogen. This assists us in determining the ideal conditions to be maintained inside the vessel at any time during the long-term storage period.

	Orthohydrogen	Parahydrogen	Normal hydrogen
C_p (J/Kg·K)	-	3.61642 [19]	3.46074 [19]
C_v (J/Kg·K)	-	-	-
μ (Pa·s)	-	7.5e-6 [20]	8.766e-6 [21]
ρ (g/cm^3)	-	0.05193 [20]	0.80844e-3 [22]
k (mW·m^{-1}K^{-1})	-	192.38 [22]	186.97 [22]
Pr	-	-	-
s (J/K)	-	14.19825 [19]	15.60877 [19]
h (KJ/mol)	-	3.38644 [19]	3.41437 [19]

TABLE 2: Properties of various states of hydrogen at room temperature (~293 K).

	Orthohydrogen	Parahydrogen	Normal hydrogen
C_p (J/Kg·K)	-	2.84128 [19]	2.59041 [19]
C_v (J/Kg·K)	-	-	-

	Orthohydrogen	Parahydrogen	Normal hydrogen
μ (Pa·s)	-	355e-3 [23]	3.416e-6 [21]
ρ (g/cm^3)	-	0.0010784 (g mol/cc) [24]	-
k (mW·m^{-1}K^{-1})	-	0.0605e3 [25]	68.62 [21]
Pr	-	-	-
s (J/K)	-	9.51828 [19]	11.68214 [19]
h (KJ/mol)	-	2.55403 [19]	4.11160 [19]

TABLE 3: Properties of various states of hydrogen at ~70 K.

Furthermore, it can be observed that the difference of thermal conductivity between ortho- and para-hydrogen in the gaseous state is ~0.5% at ~20 K. Thermal conductivity differs due to the differences in low pressure C_p of ortho- and para-hydrogen at intermediate temperatures. The ratios of those C_ps was calculated by Farkas (1935) as follows: $\frac{K_{(p)}}{K_{(n)}} = \frac{C_{v(p)}+2.25R}{C_{v(n)}+2.25R}$. Ubbink (1948) reported that he couldn't detect any differences in thermal conductivities between para- and normal-hydrogen at 17 K. However, Heizinger (1960) determined $K_{(p)}$ to be 0.57±0.07% higher than $K_{(n)}$ at 20 K. A year later he reported the difference to be a function of ortho-para hydrogen composition at 20 K [19].

T, K	Kp/Kn	T, K	Kp/Kn
10	1.000	160	1.196
20	1.000	170	1.183
30	1.000	180	1.169
40	1.001	190	1.152
50	1.004	200	1.136
60	1.015	210	1.120
70	1.035	220	1.104
80	1.065	230	1.090
90	1.100	240	1.077
100	1.135	250	1.066
110	1.165	260	1.058
120	1.187	270	1.047
130	1.200	280	1.040
140	1.206	290	1.033
150	1.203	300	1.028

TABLE 4: Calculated values of the ratio of thermal conductivity of gaseous para to normal hydrogen. As reported in [19] Hust et al. (1965).

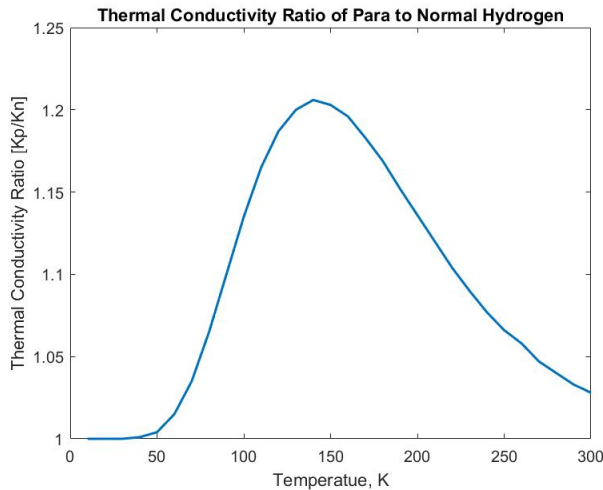**Thermal Conductivity Ratio of Para to Normal Hydrogen**

FIGURE 2: Thermal conductivity ratios of para to normal hydrogen along a range of temperatures.

There is significant difference between rotational energies of parahydrogen and orthohydrogen at temperatures below 250 K. Additionally, the maximum difference occurs around 145 K [10]. The heat capacity of a certain constant mixture of ortho- and para-hydrogen can be calculated using this equation: $C_{P(mix)} = X_{(p)}C_{P(p)} + X_{(o)}C_{P(o)}$ where X is the ratio of the component present relative to the other ($X_{(p)} = 1 - X_{(o)}$) [19]. It's worth noting that the isobaric heat capacity of equilibrium hydrogen experiences an increase of ~50% as the temperature increases from 20.4 K to 90 K [27].
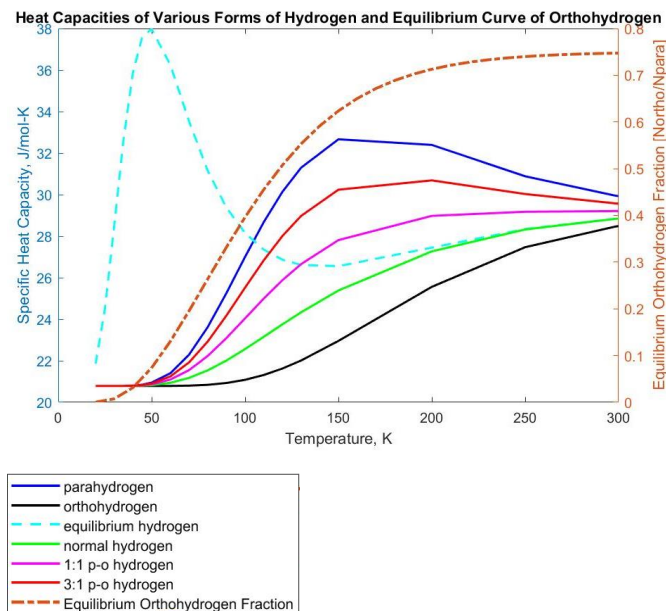
Heat Capacities of Various Forms of Hydrogen and Equilibrium Curve of Orthohydrogen

- parahydrogen
- orthohydrogen
- equilibrium hydrogen
- normal hydrogen
- 1:1 p-o hydrogen
- 3:1 p-o hydrogen
- Equilibrium Orthohydrogen Fraction

FIGURE 3: Ideal-gas heat capacities of parahydrogen, normal hydrogen, and orthohydrogen.

4. CONCLUSION

When outlet temperature of hydrogen increases, the conversion has less of an effect on the total hydrogen heat capacity. Furthermore, additional data on mechanical and thermal properties of ortho- and para-hydrogen at room temperature and at 77 K need to be collected and considered into the design. We plan on plotting thermal conductivities of ortho- and para-hydrogen over our considered range of temperatures to analyze patterns and differences. As well as look into the benefits of using an adsorption compressor and consider implementing it in our design. Also, we are in the process of creating an ANSYS simulation to demonstrate the adsorption process inside the vessel.

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