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Design and Operation of Liquid Hydrogen Storage Tanks

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Liquid hydrogen (LH2) is a versatile and efficient energy carrier with numerous applications in space exploration, hydrogen fuel cell vehicles, industrial processes, and the maritime sector. However, its extremely low boiling point and low density present unique challenges in handling, storage, and transportation, particularly in the prevention of loss of containment scenarios. At present, there is still limited knowledge available on the thermodynamics of liquid hydrogen contained in cryogenic storage tanks. This scientific paper delves into an examination of insulation techniques and the operation of liquid hydrogen tanks. Also, self-pressurization is explained and set into context. Furthermore, modelling of specific parameters such as temperature distribution, pressure increase and liquid level play an important role in understanding the thermodynamics inside of LH2 tanks and enable to draw conclusions for the efficient operation when avoiding the loss of hydrogen by releasing boil off gas. The ramifications of this study hold critical importance for industries reliant on hydrogen. The insights gained will facilitate the development of prediction models to enhance operational directives, and the development of effective storage systems.

* 1. Introduction

Hydrogen technologies are increasingly vital in transportation and industry, with safe and efficient storage and transfer methods being crucial for maximizing its energy storage potential. Hydrogen, being the lightest and most abundant element in the universe, has attracted significant attention as a possible solution to address global energy and environmental challenges. Its remarkable gravimetric energy density and eco-friendly properties as a fuel make it a promising alternative to fossil fuels and enable green energy production and transportation. Compared to conventional fuels, hydrogen boasts a higher energy content per unit mass, with a gravimetric energy density of 120 MJ/kg, surpassing gasoline's 44 MJ/kg. However, its volumetric energy density is lower, with liquid hydrogen offering 8 MJ/l compared to gasoline's 32 MJ/l (“Hydrogen Storage,” 2023). Despite its potential, ensuring efficient utilization of hydrogen remains a top priority. Thermally insulated storage tanks are essential for maintaining the cryogenic conditions required for liquid hydrogen, which is stored at -253°C close to atmospheric pressure, until further distribution or use (“liquid hydrogen – Rocketology,” 2016). Liquid hydrogen serves various applications, notably as a rocket propellant in space exploration due to its high thrust and clean combustion with water vapor as the primary byproduct (“liquid hydrogen – Rocketology,” 2016). Additionally, it shows promise as a fuel for ground vehicles, where it can be stored onboard and used in fuel cells to generate electricity. These efforts align with the goal of developing cleaner and more sustainable transportation options. However, the storage of liquid hydrogen poses obstacles due to the low temperatures and a lack of knowledge on its behavior during transfer operations between storage tanks. This paper provides an overview of different insulation materials for liquid hydrogen (LH2) tanks, along with insights into their normal operation, transfer processes, and self-pressurization mechanisms. It also explores simulation approaches, to manage boil-off gas effectively. Ultimately, the aim is to offer critical insights into the cryogenic storage of hydrogen, addressing current state-of-the-art practices and the challenges that lie ahead.

* 1. Insulation Techniques for Cryogenic Applications

Cryogenic tanks, commonly used to store substances like LH2 or liquefied natural gas (LNG), typically consist of double-walled containers. Insulation material is positioned between the outer and inner walls, the so-called annular space, under vacuum conditions to minimize heat transfer by convection. This section provides an overview of various materials and insulation concepts utilized in such tanks. Some insulation materials for the containment and storage of LH2 are:

* Multi-layer insulation (MLI)
* Perlite
* Hollow glass microspheres (HGM)
* Aerogel
* Foams
  + 1. Overview of Different Materials

Multi-layer insulation (MLI) is commonly used in mobile applications and smaller vessels. MLI is a type of insulation system that consists of multiple layers of thin reflective materials (e.g. aluminum foils or aluminized polymers) able to reduce heat transfer by radiation, separated by spacers. These spacers usually made of polymers (e.g. polyester) prevent heat transfer by conduction. Furthermore, MLI is often vacuumized to prevent heat convection. For instance, the insulation system may consist of 70 layers of aluminized polymers, interspersed with glass fibers or polymer spacers, resulting in a combined thickness of around 30 mm for mobile applications such as the automotive sector.

Perlite is a mineral known for its versatility and minimal environmental impact in mining and processing. Its utilization in various products, such as insulation, is seen as a sustainable solution, deriving from a naturally occurring material abundantly found worldwide. Subjected to controlled rapid heating, perlite ore expands, increasing up to 20 times its original volume. This expansion creates a foam-like cellular structure characterized by clusters of microscopic glass bubbles. As a result, expanded perlite exhibits exceptional insulating properties while maintaining low density. Commonly, double-walled vessels with annular spaces filled with perlite are employed for storing cryogenic fluids like liquid hydrogen and liquid helium (Perlite Institute, 2024).

Glass microspheres often refer to as “glass bubbles” represent an innovative approach to cryogenic insulation. These hollow glass microspheres (HGM) are microscopic borosilicate spheres usually with a nominal diameter of 60 microns and a bulk settled density of approximately 65 kg/m3 which effectively fill the annular space, offering superior thermal insulation properties. Crush testing, vibration tests, and thermal performance evaluations consistently affirm the superiority of glass bubbles which, unlike traditional materials like perlite powder, do not compact or break, ensuring consistent insulation performance. Despite their higher cost (up to three times compared to perlite powder), glass bubbles demonstrate enhanced heat transfer resistance, resulting in improved insulation efficiency. Field testing confirms their effectiveness, with significant reductions in boil off gas (BOG) formation observed over multiple thermal cycles. Overall, glass bubbles provide reliable and durable insulation, making them a promising choice for cryogenic applications (Fesmire et al., 2022). Field testing of a 190-m3 VJ LH2 sphere at Stennis Space Center further validates the efficacy of glass bubbles, demonstrating a remarkable 46% reduction in BOG formation over three thermal cycles in six years (Fesmire et al., 2022).

Aerogel is characterized by its extremely low density and porous structure, which impart excellent insulating properties. Additionally, aerogel is lightweight and flexible, making it suitable for use in various tank geometries and configurations. Aerogel is described as an amorphous silica and exhibits noticeable differences at a microscopic level compared to other materials. Aerogel beads are manufactured by a multitude of companies and are advertised under different names. The morphology, microstructure, and submicroscopic features of aerogel play a crucial role in determining how heat energy is transmitted through the material at specific vacuum levels (Scholtens et al., 2008).

Foams insulation is another possible component of passive thermal protection insulation technology in cryogenic applications. It works alongside materials like spray-on foam insulation (SOFI), fiber-reinforced plastic (FRP), aerogel, and HGMs within a vacuum MLI system to minimize heat transfer. Studies have highlighted the significant role of MLI in thermal protection, particularly at high vacuum levels, while foam insulation primarily contributes to insulation at atmospheric pressure (Yin et al., 2024).

* + 1. Performance of different insulation systems

In cryogenic applications, insulation materials must meet several critical criteria to ensure optimal performance and safety. Fire resistance is paramount, as insulation materials should possess properties such as being non-combustible and preserving its performance during fire, enhancing overall safety. Durability is equally essential, as insulation materials must withstand harsh environmental conditions and resist degradation over time to maintain their effectiveness and longevity. Cost considerations play a significant role in decision-making, both in terms of initial investment and long-term cost-effectiveness. While upfront costs are important, evaluating the overall cost-effectiveness of insulation materials over their lifespan is crucial, taking into account factors such as maintenance requirements and potential energy savings. Effective thermal conductivity is a fundamental characteristic that directly impacts insulation performance. Materials with lower effective thermal conductivity are preferred, as they effectively limit transfer of heat, helping to maintain stable temperatures and minimize heat loss in cryogenic environments. Table 1 gives an overview.

*Table 1: Comparison of different insulation materials (Fesmire et al., 2022; Fesmire, 2015; HEATERK, 2022)*

|  |  |  |  |
| --- | --- | --- | --- |
| Insulation material | Effective thermal conductivity (W/m\*K) | Density (kg/m3) | Melting temperature (℃) |
| Vacuum-multi-layer 1 (polyester) | 10-6 to 10-5 | 42 | 140-400 |
| Vacuum -multi-layer 2 (fiberglass) | 10-6 to 10-5 | - | 1000-1400 (660 for Aluminium) |
| Glass bubbles | 10-3 to 10-4 | 65 | 1400-1600 |
| Perlite | 10-4 to 10-3 | 132 | 1260 |
| Aerogel Blanket | 10-3 to 10-2 | 133 | 1200 |

The prevalent insulation options for LH2 tanks include perlite and MLI, with NASA recently opting for glass bubbles. NASA's latest storage tank combines two innovative technologies to enhance large-scale LH2 storage and control capabilities, merging active and passive thermal control methods. The new NASA tank features an evacuated insulation system utilizing HGMs (Fesmire et al., 2022). Developed over the past twenty years at the Cryogenics Test Laboratory at NASA Kennedy Space Center, this insulation system has demonstrated superior thermal performance, mechanical integrity, and vacuum resilience. Complementing this passive thermal control system is an active thermal control mechanism, comprising an internal heat exchanger designed to accommodate the Integrated Refrigeration and Storage (IRAS) system. This heat exchanger, situated within the inner vessel, facilitates heat dissipation from the bulk liquid by connecting it to a refrigerator circulating cold helium gas. Upon full implementation, the IRAS system will enable precise storage management, including control of pressure, elimination of boiloff, and potentially the production of subcooled LH2 for future launch vehicles (Fesmire et al., 2022).

* 1. Liquid Hydrogen Tank Dynamic
     1. Operation of Liquid Hydrogen Tanks

LH2 terminals encompass a multitude of components crucial for efficient and safe operation such as transfer systems (comprising transfer lines, valves, and piping, which may incorporate cryogenic pumps), safety devices, control systems, and monitoring systems. These critical components bear the potential for malfunctioning which may lead to system failures. The temperature contrast between the stored liquid and the surrounding environment inevitably leads to heat transfer into the LH2, resulting in its evaporation and generation of BOG. This contributes to an increase in pressure within the storage tank, a process termed self-pressurization. To mitigate pressure buildup, venting BOG from the tank into the atmosphere thanks to the opening of a pressure relief valve (PRV) is necessary, resulting in the loss of valuable hydrogen. By releasing the content of the tank, the temperature remains constant. This process is called auto-refrigeration (Verfondern, 2008). The generation rate of BOG fluctuates based on factors like the quality of insulation, the surface-to-volume ratio of the tank, and its capacity. For instance, a 50 m3 cryogenic tank may experience BOG generation on the order of 0.4% per day, whereas a larger 20,000 m3 LH2 tank may experience a lower rate of around 0.06% per day (Verfondern, 2008). Furthermore, the ingress of heat causes the liquid in contact with the inner wall of the tank to heat up and therefore expand. The warmer, lighter liquid is then ascending to the vapor-liquid interface. This way a local heat convection is initiated, that promotes thermal stratification (Kang et al., 2018) of the liquid phase and accelerates pressurization. Heat conduction from the warmer interface contributes to increase the temperature the warmer liquid layer. The phenomenon of self-pressurization holds significant importance in ensuring the efficient storage of LH2. On the other hand, it becomes necessary to release the BOG when the pressure within the storage tank reaches a certain threshold to prevent overpressure and potential rupture of the container. Such an eventuality could lead to catastrophic consequences due to loss of containment. Therefore, the design of the tank plays a critical role in determining the opening pressure of the PRV. Additionally, understanding the self-pressurization dynamics of the tank is vital to prevent hydrogen loss through BOG, which equates to a loss of stored or transported energy. Self-pressurization can be exploited to extract hydrogen from the tank for conversion into electricity via a fuel cell or combustion in an internal engine. Moreover, predictive pressure adjustment techniques can be employed to establish optimal conditions to bridge limited time periods when economical hydrogen utilization is not feasible. Figure 1 shows a schematic of a land based LH2 tank.

Diagram of a diagram of a circle with arrows and a circle with red arrows

Description automatically generated

*Figure 1: Schematic of a LH2-cryotank with heat and BOG fluxes*

* + 1. Modelling of liquid hydrogen tank dynamic

Modeling the pressurization process remains essential for gaining a comprehensive understanding of the phenomenon. This section will provide a concise overview of modeling approaches. Several numerical models including computational fluid dynamics (CFD) have been developed specifically for cryogenic fluids like liquefied natural gas (LNG). These models span from those concentrating on heat ingress and consequent BOG formation rates to those examining heat transfer between vapor and liquid phases. Agrawal et al., (2017) worked on a model for two-phase thermal stratification to simulate the accumulation of stratified mass in a cryogenic tank. The model was rigorously validated. Subsequent parametric studies were conducted to examine the impact of varying liquid sub-cooling on the evolution of stratified layers and mass within the tank. Results indicated that stratification grows more rapidly in tanks containing cryogenic liquid with higher levels of sub-cooling. The study concluded that tanks with higher liquid sub-cooling and pressure tend to accumulate more stratified mass. Consequently, it was recommended that cryogenic fluid systems should be designed to operate at the lowest feasible pressure to minimize the accumulation of stratified mass and the amount of unusable propellant in the tank, thereby optimizing payload efficiency (Agrawal et al., 2017). Joseph et al., (2017) developed a transient two-phase thermodynamic lumped model to simulate thermal stratification in a typical LH2 tank. The tank has specific dimensions, with the focus being on the thickness of the foam insulation, with the liquid level filled up to 87% of the total height. The model simplifies the complex two-dimensional boundary layer phenomenon into a one-dimensional lumped model to predict pressure evolution and bulk fluid temperatures during thermodynamic transients in the tank. The computational domain includes discretization of tank surfaces and the insulation as nodes, and bulk fluid as lumps. Boundary flow zones near the tank wall contribute to heat in-leak and boundary layer flow, while a core zone experiences negligible flow and relies on fluid conduction for heat transfer. The study defines stratified mass as the total mass of fluid with temperatures above the pump cavitation limit (Joseph et al., 2017). A model, developed by Ustolin et al., (2021), aims to replicate fire tests conducted by BMW as part of the hydrogen 7 project. This model, designed as a lumped model, is intended for analyzing the thermal response of cryogenic tanks. It adopts a thermal nodes approach, partitioning the domain into eight nodes to comprehensively address various aspects: the liquid and vapor phases, sections of the inner shell in contact with the liquid and vapor, insulation above and below the liquid level, and segments of the external tank wall above and below the liquid level. In this model, considerations encompass heat and material balances on the vessel subjected to fire, encompassing phenomena such as boiling regime, heat-up, and pressure build-up in the cryogenic tank. Mass balances for liquid and gaseous hydrogen, thermal balances for all nodes, tank pressure, and LH2 level are determined using a system of partial differential equations. To replicate hydrogen evaporation and condensation, the model integrates non-equilibrium conditions, wherein liquid and gaseous phases exhibit distinct temperatures despite sharing the same pressure. It employs the Knudsen model to compute hydrogen evaporation and condensation mass fluxes, alongside equations to estimate convective heat transfer coefficients. Furthermore, the model accommodates the pressure relief valve (PRV) effect, considering both natural and forced convection during PRV closure and opening, respectively. It assesses hydrogen mass flow rate during PRV activation and incorporates radiative heat flux between the inner vessel wall and the liquid, as well as the thermal load from the engulfing fire. Ultimately, the model offers a conservative estimate of the time to failure (TTF) by positing container rupture when the mechanical stress from pressure build-up equals the yield strength of the shell material (Ustolin et al., 2021).

Furthermore, Ustolin et al., (2022) conducted a two-dimensional computational fluid dynamics (CFD) analysis to examine the thermal behavior of a cryogenic liquid hydrogen (LH2) vessel exposed to fire. The analysis incorporates the evaporation and condensation of the LH2, enabling the prediction of tank pressurization rate and temperature distribution. The study assumes the complete engulfment of the vessel in the fire as a worst-case scenario. Validation of the CFD model was achieved through comparison with the results of a small-scale fire test on an LH2 tank. The findings offer crucial insights into the dynamic response of the cryogenic tank in such a worst-case accident scenario. Utilizing the tank pressurization and temperature distributions from the case study can provide conservative estimates of the time to failure (TTF) of the vessel (Ustolin et al., 2022).

Matveev and Leachman, (2023) discussed the importance of accurately predicting hydrogen evolution within storage tanks, focusing on pressurization rates and necessary venting rates. The analysis employs a lumped-element approach, emphasizing the impact of tank size on storage performance. This method allows for quick studies during early design stages, avoiding the high costs of CFD simulations. The study utilizes an aluminum tank resembling those used in space travel applications, extensively tested by NASA, to explore the scaling aspect of tank storage capabilities. The main objective is to demonstrate the efficacy of the lumped-element modeling method in assessing the effects of tank size on self-pressurization processes and required venting rates, essential for practical tank users conducting trade-off studies considering space, cost, and complexity constraints. The model's versatility allows for its application in various operational regimes such as liquid supply/extraction and cryocooling (Matveev and Leachman, 2023).

Table 2 gives an overview on the described modeling approaches, their focus and validation.

*Table 2: Overview of existing models for thermodynamics behaviour of cryo-tanks)*

|  |  |  |  |
| --- | --- | --- | --- |
| Model | Model Focus | Validation | Modelling Approach |
| Kassemi et al. (2014) | Self-pressurization of large volume LH2 storage tank | Experimental testing with  a multipurpose hydrogen test bed (MHTB) | CFD model utilizing kinetics-based Schrage equation |
| Stewart et al. (2016) | Self-pressurization | Comparison with experimental data for pressure evolution and temperature | CFD simulation in ANSYS Fluent |
| Agrawal et al. (2017) | Thermal stratification | Experimental data from literature | - |
| Joseph et al. (2017) | Thermal stratification | - | Transient two-phase thermodynamic lumped mode |
| Ustolin et al. (2021) | Thermal behavior of LH2 tank when exposed to fire | Experimental validation | Lumped-element model |
| Ustolin et al. (2022b) | Thermal behavior of LH2 tank when exposed to fire | Small scale fire test on LH2 tank | Two-dimensional CFD analysis |
| Matveev and Leachman (2023) | Pressurization- and venting rates | - | Lump-element models |

* 1. Conclusions

Cryogenic tanks for LH2 or LNG rely on insulation materials under vacuum conditions to minimize heat transfer and maintain stable temperatures and pressures in time. Materials such as MLI, perlite, HGM, aerogel, and foam insulation play crucial roles in these applications. Considerations such as fire resistance, durability, cost-effectiveness, and thermal conductivity are pivotal in selecting the most suitable insulation material for cryogenic tanks. Additionally, the operation of LH2 tanks, delving into self-pressurization dynamics and modeling techniques was outlined. Challenges arise from self-pressurization, driven by heat transfer, leading to BOG generation and eventually hydrogen loss. Modeling plays a key role in understanding and predicting these processes. Various numerical and analytical models were developed for cryogenic fluids, addressing different aspects such as heat ingress and BOG rates. These models contribute significantly to understanding and optimizing the storage and handling of cryogenic fluids, particularly LH2. While existing models offer insights into specific scenarios, to date there is no specialized model available that is capable of predicting the pressure increase in LH2 tanks for different types of tanks with a low computing cost. Therefore, there's a growing demand for adaptable models applicable to various LH2 containers under different operating conditions. Such endeavors will be crucial for advancing LH2 storage technology and bolstering the development of a hydrogen-centric energy infrastructure.

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