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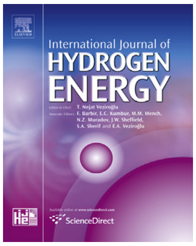

Development of a small-scale hydrogen liquefaction system

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ABSTRACT

A liquid hydrogen (LH_2) storage tank, vacuum jacketed transfer line, and hydrogen liquefier have been designed, built, and coupled together to form a small scale hydrogen liquefaction plant. The liquefier was designed to liquefy at 1 L/hr at 3 bar with a single stage Gifford-McMahon cryocooler. The liquefier can hold up to 200 L of LH_2 in a multi-layer insulated (MLI) vacuum jacketed storage tank. The liquefier includes two ortho-para hydrogen (O–P) converters, a liquid nitrogen precooler and a heat pipe. After liquefaction, LH_2 can be transferred to a 5 L storage vessel using a flexible, MLI and vacuum insulated low loss transfer line. This storage vessel was designed to limit boil-off by using two 1 m long G10-CR necks to reduce conduction, a radiation shield and MLI with high vacuum to reduce radiation and convective heat leak from the outer shell to the inner tank. Presented in this paper are more detailed system configurations and results of various operational modes and applications.

Introduction

With the recent publishing of the fifth assessment report on climate change by the Intergovernmental Panel on Climate Change (IPCC) [1], it has become clearer that the world needs to focus on continuing the development of clean technologies and energy storage systems to address global climate change. Hydrogen used for energy storage offers one of many solutions to help reduce our carbon foot print. Hydrogen is currently produced from several methods such as steam methane reforming or electrolysis [2]. Storing energy in the form of hydrogen is one method that can help address the growing problems facing this world related to our growing energy consumption. There are various methods of storing hydrogen including compression, metal hydrides, storage as a cryogenic liquid or a combination. Storage of hydrogen as a liquid offers a low pressure high energy density fuel that can be used in a variety of applications [3–6]. In the past, liquid hydrogen (LH_2) has been most widely known for its use as a rocket fuel [7]. Today, there are many applications emerging for this high energy density fuel such as development of fuel cell vehicles using cryo-compressed hydrogen storage tanks by Lawrence Livermore National Laboratory, BMW, and Linde

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Group [6]. While hydrogen's use in vehicles may or may not be the most practical option, hydrogen does offer niche applications, such as aviation, a very efficient light weight fuel for operations [8]. Boeing and The Office of Naval Research are using liquid hydrogen as the fuel for unmanned aerial vehicle (UAV) development [9,10]. With the growing interest in hydrogen, there has been various research conducted on small scale hydrogen liquefaction systems [11,12]. Previous research successfully carried out at the Korean Institute of Science and Technology (KIST) in 1997 developed a small scale liquefier and storage tank [13]. This paper describes the design, fabrication and operation of a new 1 L/hr at 3 bar absolute (bara) hydrogen liquefier and a separate 5 L storage tank to further improve hydrogen storage technology. The two systems demonstrated liquefaction, transfer and re-liquefaction of hydrogen to create a minimal loss hydrogen storage system.

Component design and fabrication

Liquefier

Thermal requirements and cryocooler selection

A liquefier capable of 1 L/hr liquefaction at 3 bara, also capable of storing up to 200 L of LH_2 was designed, fabricated and demonstrated. Using NIST's thermodynamic property data for hydrogen, including heat of conversion (516 kJ/kg) from ortho-hydrogen to para-hydrogen (O–P), 80 W of cooling would be needed to achieve 1 L/hr liquefaction [14]. With this data, a market search of available cryocoolers was conducted; cooling 70 W at 20 K or 100 W at 25 K, a model AL325 GM cryocooler from Cryomech Inc. was selected [15].

Structural design and heat leak mitigation

To limit heat load on the system, similar to other conventional cryogenic storage systems, this liquefier used a double walled vacuum jacketed Dewar design [16]. In this system, there are two concentric tanks, an inner tank and outer tank, made of 304 stainless steel (304 SUS). Between the two tanks is multilayer insulation (MLI) and high vacuum of 10^{-4} ~10^{-5} Torr. Forty layers of Lydall's double sided aluminized Mylar (DAM) with CRS-wrap (a micro-glass spacer) were used to wrap all surfaces of the inner vessel, Fig. 1 [17]. A nEXT400D turbomolecular pump (TMP) from Edwards with a rotary vane backing pump was used in this experiment. A series of G-10 CR baffles wrapped in 5 layers of DAM without the spacer were fabricated to reduce convective and radiative heat transfer down through the inner tank's neck. To reduce conductive heat transfer to the inner tank, large G-10 CR disk was used between the outer tank and the inner tank. Finally, to reduce the stress on the inner tank's neck from its weight, a low loss G-10 CR support structure was manufactured and installed at the bottom between the inner and outer tanks. After taking all these steps to reduce heat leak, a calculated 10 W of heat leak was expected.

Hydrogen heat pipe and ortho-para converter

AL325 cold head has limited surface area and it is situated at the top of the liquefier; to quickly transfer heat from the lower sections of the tank to the cold head and to improve surface area for liquefaction, the fabrication of a heat pipe would be necessary. The heat pipe body was fabricated from a 304 SUS pipe. Two oxygen free high conductivity (OFHC) caps were machined with extended concentric fins on one side and a smooth mirror finish on the other. The fins are for increased surface area in the inside of the heat pipe and the mirror finish is to reduce contact resistance between the cold head or O–P converter and the copper caps. After machining, the caps were silver brazed to the top and bottom of the heat pipe. To further improve external surface area, copper axial fins were welded to the outer surface of the pipe. The body was pressurized with helium to 36 bara for leak testing. There was also an O–P converter mounted to the bottom OFHC block of the heat pipe. The converter was a stainless lower cylinder with an OFHC block cap which had been silver brazed together. Hydrogen flow would enter into the converter through a flow path in the copper block cap where it would cool or condense and then flow down onto a commercial 30–50 mesh iron (III) oxide ($HFeO_2$) catalyst bed. After converting in the bed, the hydrogen would flow out through the bottom of the bed into the main storage tank. Storage as para hydrogen is especially important when transferring LH_2 to external unrefrigerated tanks because the heat of conversion causes increases boil-off [16]. This O–P converter ensured that over 99% all the liquid hydrogen produced was para hydrogen.

Precooler

To improve liquefaction rates, a precooler was also fabricated. The use of a liquid nitrogen (LN_2) precooler with O–P catalyst significantly reduced the heat load on the cryocooler. The sensible heat of hydrogen between 300 K and 20 K is 3509 kJ/kg and 2905 kJ/kg between 300 K and 77 K [14,16]. Therefore, 83% of this heat can be removed by using a precooler. The system was designed as a packed bed heat exchanger that was placed in a commercial 35 L LN_2 Dewar. For the design, hydrogen gas at ambient temperature would be first flowed through a vacuum shielded pipe; this initial vacuum insulation would prevent impurities from condensing before reaching the activated carbon. In the upper section of the packed bed, there was approximately 400 mL of 1–2 mm (10–18 mesh) activated charcoal to help reduce any impurities in the gas. Next, the gas would pass through approximately 100 mL of the commercial O–P iron (III) oxide catalyst in the bottom of the bed. After passing through the catalyst, the gas would then pass back through an uninsulated line to remove any additional heat from O–P conversion and then would enter a vacuum jacketed bayonet adaptor for the transfer line.

Transfer lines and bayonets

Several bayonets with valves and two 2 m flexible vacuum jacketed transfer lines were fabricated. These transfer lines connects the precooler to the liquefier during liquefaction and were reconfigured between the 5 L and liquefier during liquid transfer and re-condensation. For temperature measurement, one of the transfer lines was outfitted with E-type thermocouples welded directly to its inner pipe surface. After installing the thermocouples, DAM MLI was wrapped over the