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Characterization of leaks from compressed hydrogen dispensing systems and related components

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Abstract

The equations are developed for the calculation of leak flow rates in various leak regimes. Leaks due to pressure-driven convection and due to permeation through metals are considered. For convective leaks, the conditions under which the flow transitions from laminar to turbulent and from subsonic to choked (sonic) flow are discussed. Equations are presented to calculate leak rates for subsonic laminar and turbulent flows, as well as choked (sonic) flow rates. Given the advantages of using noncombustible gases for leak testing and measurement, equations are also developed for calculating the equivalent leak rate of helium when it is used as a surrogate for the combustible gases hydrogen and methane in each of these flow regimes. Equations are derived for the permeation rate of hydrogen through several common metals. Tabulated data is presented for the permeation rates of hydrogen through pure iron and two types of stainless steel over a pressure range from 5000 to 15,000 psi and a temperature range of −40–100 °C. The results clearly show the sensitivity of flux to temperature, with over an order of magnitude increase in flux as the temperature is increased from ambient to 373 K (100°C). Permeation rates are also found to vary significantly with material. For example, permeation rates for construction steel (as estimated from pure iron) are about three orders of magnitude higher than 403 stainless steel and nearly five orders of magnitude higher than type 316L stainless steel for a given temperature and pressure. Under many combinations of pressure and temperature, leak rates for Fe exceed the permissible gaseous hydrogen leak rates, while rates for 316L stainless steel are well below permissible permeation rates at all combinations of temperature and pressure considered.
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1. Introduction

The system components expected to evolve during the future development of a hydrogen infrastructure include storage, bulk transportation and distribution, production and utilization. One common requirement that must be met in each of these components is the safe confinement and utilization of hydrogen. The identification of critical safety issues thus becomes integral to hydrogen infrastructure development. To this end a recent workshop was held to identify safety scenarios and prioritize research and technical activities to support the development of hydrogen safety standards [1]. As part of this workshop, unintended releases of hydrogen were recognized as a critical safety issue that needs to be addressed. Of the 68 release scenarios identified, the majority were leaks that resulted in combustible cloud formation. The leak size, its origin and surroundings

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were all used to further categorize the scenarios and resulting hazards.

It is generally recognized that two categories of leaks could be important in future hydrogen infrastructure components. The first of these is the leakage of gaseous hydrogen through holes, breaks and defects in material surfaces that contain the hydrogen. These leaks involve the convection of hydrogen through openings of various size and are driven by a pressure difference. They fall in the continuum flow regime, as opposed to the molecular flow regime, and involve openings that are many times the molecular mean free path. Gaseous convective leaks can further be divided into leaks from high-pressure sources and those from low-pressure sources, since the pressure ratio across the leak determines whether the flow through the leak is subsonic, or choked (sonic). For example, in the simple case of air passing through a small nozzle, if the pressure ratio across the nozzle is greater than about 1.89:1 the flow will be choked at the nozzle exit and the flow rate through the nozzle will be limited. In this case the flow is considered compressible and the equation set governing compressible flow behavior can be applied. At pressure ratios across the leak that are below the choked flow minimum, the flow is subsonic and governed by incompressible flow equations. Depending on leak geometry and flow rate, this subsonic flow can be either laminar or turbulent.

A second leak category is diffusion, or permeation, of hydrogen through the surface of the hydrogen confinement vessel. In this case the leak falls into the molecular flow regime and involves the diffusive transport of molecules through the surface material. This type of leak is probably most relevant to storage tanks that have a high surface area and long residence times where the leak occurs over an extended period of time. Again, this type of leak is governed by a different set of mathematical models.

Hydrogen system components will require periodic testing as to their integrity against leaks of various types. From the standpoint of both safety and cost, it is desirable to test for leaks and assess whether the leak rate is below a desired minimum standard from various components using an inert surrogate gas. Helium has been proposed as a good candidate for this procedure due to its low molecular weight and molecule size, which are comparable to, but not exactly matching, those of hydrogen.

The objective of this paper is twofold. The first objective is to provide a framework for the calculation of flow rates in various leak regimes. As such, the paper addresses both convectively driven gaseous hydrogen leaks and the permeation of hydrogen through metals. In the case of convective leaks, equations will be presented for determining leak rates for low-pressure (laminar and turbulent incompressible flow) and high-pressure (choked flow) leaks. Given the desirability of using an inert gas such as helium as a surrogate for combustible gas during leak testing. A second objective is to develop the equations for calculating the equivalent flow rate of helium when it is used in leak testing as a surrogate for two common combustible gases, hydrogen and methane. In the following sections convective leaks are first considered, followed by a discussion of hydrogen permeation through metals. Convective leaks are further categorized into subsonic laminar and turbulent flows, and choked flows from high-pressure sources. In each case the governing equations are developed and used to determine the equivalent flow rates of a helium surrogate gas that replaces the hydrogen and methane.

2. Hydrogen leakage

Gas leakage from gas storage and delivery systems typically occurs through an aperture that is much greater than the mean free path of the gas molecules. Therefore, a flow-based continuum model can be used to approximate the leakage rate. The highest pressures encountered typically range from 3600 to 10,000 psi in storage tanks and 7500–15,000 psi in the fueling system lines. However, downstream in a vehicle fuel system the actual pressure can be as low as 10–15 psi g.

Swain and Swain [2] studied the leakage rates of both hydrogen and methane at line pressures up to about 14 psi. They determined that leaks at low pressures occur primarily in a laminar mode, although entrance effects must be accounted for in leaks with large cross-sections [2]. Leaks at higher pressures occur in a turbulent regime, and leaks at very high pressures can be sonic [3]. The maximum allowable leak flow rates specified in NGV2 [4] and HGV2 [5] for natural gas and hydrogen, respectively, are given in the first column of Table 1. Shown in the remaining columns of Table 1 are the calculated equivalent helium leak rates for methane and hydrogen in different flow regimes based on the procedures described below.

2.1. Determination of flow regime

At lower pressures the flow can be treated as incompressible and can be either laminar or turbulent. The transition from laminar to turbulent flow should occur at approximately the same Reynolds numbers ($Re =$