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EFFECTS OF HYDROGEN GAS ON STEEL VESSELS AND PIPELINES

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1. Introduction

Carbon and low-alloy steels are common structural materials for high-pressure hydrogen gas vessels and pipelines. These steels are low cost, and a wide range of properties can be achieved through alloying, processing, and heat treatment.[1] Fabricating complex structures such as gas containment vessels and pipelines is readily accomplished with steels since these materials can be formed, welded, and heat treated in large sections.

The containment and transport of high-pressure hydrogen gas in steel structures presents a particular challenge. Hydrogen gas can adsorb and dissociate on the steel surface to produce atomic hydrogen.[2,3] The subsequent dissolution and diffusion of atomic hydrogen into steels can degrade mechanical properties, a phenomenon generally referred to as hydrogen embrittlement. The manifestation of hydrogen embrittlement is enhanced susceptibility to fracture. Hydrogen reduces typical measures of fracture resistance such as tensile strength, ductility, and fracture toughness, accelerates fatigue crack propagation, and introduces additional material failure modes.[3] In particular, steel structures that do not fail under static loads in benign environments at ambient temperature may become susceptible to time-dependent crack propagation in hydrogen gas.

The objective of this chapter is to provide guidance on the application of carbon and low-alloy steels for hydrogen gas vessels and pipelines, emphasizing the variables that influence hydrogen embrittlement. Section 2 reviews published experience with hydrogen gas vessels and pipelines. Industrial gas and petroleum companies have successfully used carbon and low-alloy steels for hydrogen gas containment and transport but only within certain limits of material, environmental, and mechanical conditions.[4-6] In the proposed hydrogen energy infrastructure, it is anticipated that hydrogen gas vessels and pipelines will be subjected to operating conditions that are outside the windows of experience. Thus, section 4 will demonstrate trends in hydrogen embrittlement susceptibility for steels as a function of important material, environmental, and mechanical variables. The metric for hydrogen embrittlement susceptibility is based on fracture mechanics properties. Fracture mechanics principles are reviewed in section 3.

This chapter focuses on effects of hydrogen gas on steel structures at near-ambient temperatures. For these conditions, atomic hydrogen is in solid solution in the steel lattice and can facilitate fracture through one of several broadly accepted mechanisms.[7,8] Excluded from this chapter are references to hydrogen embrittlement mechanisms that are promoted by elevated temperatures or aqueous environments. A well-known mechanism in this category is "hydrogen attack", which involves a chemical reaction between atomic hydrogen and carbon in steel to form methane gas. The formation of high-pressure methane gas in internal fissures and depletion of carbon from the steel enable material failure.[3] Other mechanisms not referenced in this chapter involve the internal precipitation of high-pressure hydrogen gas.[3] Failure caused by the internal formation of methane or hydrogen gas is not considered pertinent to steel structures used in the containment and transport of high-pressure hydrogen gas.[5]

This chapter is not intended to provide detailed guidance on the design of hydrogen gas vessels and pipelines. General design approaches for structures in hydrogen gas as well as details on vessels and pipelines are available.[4,5,9,10] While this chapter emphasizes hydrogen embrittlement of steels, it does not represent a comprehensive review of the subject. The literature on hydrogen embrittlement of steels is extensive, e.g., Refs. [11-15], and includes numerous review articles.[3,16-18] The content of this chapter does complement previous publications that address hydrogen compatibility of structural materials for hydrogen energy applications.[9,19-21] Finally, while this chapter presents some specific data to illustrate hydrogen embrittlement trends in steels, the document is not intended to serve as a data archive. Such a data compilation has been created to guide the application of materials in a hydrogen energy infrastructure.[22]

2. Review of Hydrogen Gas Vessels and Pipelines

This section summarizes the experience of industrial gas and petroleum companies with steel hydrogen gas vessels and pipelines. Extensive information is published in two European Industrial Gases Association (EIGA) documents, which were created to provide guidance on the design of hydrogen gas vessels and pipelines.[4,5] The document on hydrogen gas pipelines[5] was developed jointly with the Compressed Gas Association (CGA) and has been published concurrently as the CGA document G-5.6. Presentations from a workshop sponsored by the U.S. Department of Energy[6] served as additional sources of information on hydrogen piping systems. From this collective published information, the material, environmental, and mechanical conditions that have been identified by industrial gas producers and consumers to impact performance of steel hydrogen gas vessels and pipelines are reported below.

2.1. Hydrogen Gas Vessels

The information reported here is for cylindrical and tube-shaped steel vessels, where the primary function of the vessels is to distribute hydrogen gas.[4] Current European hydrogen gas distributors have several hundred thousand vessels in service, which supply up to 300×10^6 m^3 of hydrogen gas to customers annually. Over the past two decades, these hydrogen gas vessels have functioned safely and reliably.

Failures of hydrogen gas vessels have been encountered in Europe, particularly in the late 1970s.[4] Subsequent studies of hydrogen gas vessels led to the conclusion that failures were ultimately enabled by hydrogen-enhanced fatigue crack propagation from surface defects.

2.1.1. Material Conditions Affecting Vessel Steel in Hydrogen

Experience indicates that failure of hydrogen gas vessels has been governed primarily by properties of the steel, particularly strength and microstructure.[4] These variables affect the susceptibility of the steel to hydrogen embrittlement.

The published experience for reliable hydrogen gas vessels pertains to a narrow range of steel conditions.[4] Hydrogen gas vessels in Europe are fabricated from steel designated 34CrMo4. The steel composition (Table 1) is distinguished by the alloying elements chromium and molybdenum and the concentration of carbon.

The 34CrMo4 steels are processed to produce a "quenched and tempered" microstructure. The heat treatment sequence to produce this microstructure consists of heating in the austenite phase field, rapidly cooling (quenching) to form martensite, then tempering at an intermediate temperature.[1] For hydrogen gas vessels, the heat treatment parameters are selected to produce a uniform tempered martensite microstructure and to limit tensile strength (σ_{uts}) below 950 MPa.[4]

Vessels used for hydrogen gas distribution are seamless, meaning the vessel body is fabricated without welds. Hydrogen gas vessels are ideally seamless since welding alters the desirable steel microstructure produced by quenching and tempering and introduces residual stress. Welds in high-pressure hydrogen gas vessels fabricated from low-alloy steels have contributed to hydrogen-assisted cracking.[23]

2.1.2. Environmental Conditions Affecting Vessel Steel in Hydrogen

The severity of hydrogen embrittlement in steel is affected by gas pressure, since this variable dictates the amount of atomic hydrogen that dissolves in steel.[17] Working pressures for steel vessels in hydrogen distribution applications are typically in the range 20 to 30 MPa.[4]

The inner surface of hydrogen gas vessels is susceptible to localized corrosion due to impurities that can exist in the steel and hydrogen gas.[4] Interactions between localized corrosion and hydrogen embrittlement have not been specified, however, impurities in the gas and steel are known to affect hydrogen embrittlement as described in section 4.

2.1.3. Mechanical Conditions Affecting Vessel Steel in Hydrogen

In addition to gas pressure, hydrostatic tensile stress increases the hydrogen concentration in metals.[18] This leads to high, localized concentrations of atomic hydrogen at stress risers, such as defects, thus promoting hydrogen embrittlement. Defects can form on the inner surface of hydrogen gas vessels from manufacturing or during service. One manifestation of defects that forms during service is localized corrosion pits.[4]

One of the detrimental mechanical loading conditions for steel hydrogen gas vessels is cyclic stress, which drives fatigue crack propagation.[4] Pressure cycling results from filling and emptying vessels during service. The presence of surface defects influences the mechanical conditions in the steel vessel wall. Surface defects intensify local stresses, which provide the mechanical driving force for fatigue crack propagation and concentrate atomic hydrogen in the steel. Cracks propagate by hydrogen embrittlement acting in concert with cyclic stress. After a certain number of vessel filling-emptying cycles, fatigue cracks reach a critical length. Then the cracks can extend by hydrogen embrittlement mechanisms that operate in a filled hydrogen vessel under static pressure.

2.2. Hydrogen Gas Pipelines

The information summarized here is for steel transmission and distribution piping systems that carry hydrogen gas. The industrial gas companies have accumulated decades of experience with hydrogen gas transmission pipelines and currently operate over 1000 miles of pipeline in the United States and Europe.[6] These pipelines have been safe and reliable for specific ranges of material, environmental, and mechanical conditions.

2.2.1. Material Conditions Affecting Pipeline Steel in Hydrogen

Although steel pipelines have been operated safely with hydrogen gas, specific limits have been placed on properties of the steels. In particular, relatively low strength carbon steels are specified for hydrogen gas pipelines.[5] Examples of steels that have been proven for hydrogen gas service are ASTM A106 Grade B as well as API 5L Grade X42 and API 5L Grade X52.[5,6] The compositions of these steels are provided in Tables 2 and 3. The API 5L steels containing small amounts of niobium, vanadium, and titanium are referred to as "microalloyed" steels. Microalloyed X52 steel has been used extensively in hydrogen gas pipelines.[5]

Steels for hydrogen gas pipelines are processed to produce uniform, fine-grained microstructures.[5] A normalizing heat treatment can yield the desired microstructure in conventional steels. A typical normalizing heat treatment consists of heating steel in the austenite phase field followed by air cooling.[1] A more sophisticated process of hot rolling in the austenite-ferrite phase field is used to manufacture fine-grained microalloyed steels.[1]

Material strength is an important variable affecting hydrogen embrittlement of pipeline steels. One of the principles guiding selection of steel grades and processing procedures is to limit strength. The maximum tensile strength, σ_{uts}, recommended for hydrogen gas pipeline steel is 800 MPa.[5]

The properties of welds are carefully controlled to preclude hydrogen embrittlement. One of the important material characteristics governing weld properties is the carbon equivalent (CE). The CE is a weighted average of elements, where concentrations of carbon and manganese are significant factors.[5] Higher values of CE increase the propensity for martensite formation during welding. Non-tempered martensite is the phase most vulnerable to hydrogen embrittlement in steels.[9,21] Although low values of CE are specified to prevent martensite formation in welds,[5] these regions are often still harder than the surrounding pipeline base metal. The higher hardness makes welds more susceptible to hydrogen embrittlement. The maximum tensile strength for welds is also recommended as 800 MPa.

2.2.2. Environmental Conditions Affecting Pipeline Steel in Hydrogen

Similar to hydrogen gas vessels, the hydrogen embrittlement susceptibility of pipeline steels depends on gas pressure. Industrial gas companies have operated steel hydrogen pipelines at gas pressures up to 13 MPa.[6]

Hydrogen gas pipelines are subject to corrosion on the external surface. While corrosion damage has created leaks in hydrogen gas pipelines,[5,6] interactions between corrosion and hydrogen gas embrittlement have not been cited as concerns for pipelines.

2.2.3. Mechanical Conditions Affecting Pipeline Steel in Hydrogen

Hydrogen gas transmission pipelines are operated at near constant pressure,[5,6] therefore cracking due to hydrogen embrittlement must be driven by static mechanical forces. Cyclic loading, which can drive fatigue crack propagation aided by hydrogen embrittlement, has not been a concern for hydrogen gas transmission pipelines.[5] Experience from the petroleum industry, however, has demonstrated that hydrogen-assisted fatigue is possible with hydrogen gas distribution piping.[6]

Defects can form on the inner and outer surfaces of steel pipelines from several sources, including welds, corrosion, and third-party damage.[5,6] Welds are of particular concern since steel pipelines can require two different welds: longitudinal (seam) welds to manufacture sections of pipeline and girth welds to assemble the pipeline system. These welds are inspected to detect the presence of defects. Similar to hydrogen gas vessels, defects in pipeline walls intensify stresses locally, creating more severe mechanical conditions for crack extension and concentrating atomic hydrogen in the steel.

3. Importance of Fracture Mechanics

Experience has revealed that defects can form on the surfaces of both hydrogen gas vessels and pipelines.[4,5] Since elevated stresses arise near defects in pressurized vessels and pipelines, establishing design parameters based on average wall stresses and material tensile data (i.e., strength and ductility) can be non-conservative. The design of structures containing defects is more reliably conducted using fracture mechanics methods. The application of fracture mechanics to structures exposed to hydrogen gas has been well documented.[3,7,9,10]

Fracture mechanics methods are commonly implemented in materials testing protocols. Fracture mechanics-based material properties are needed for engineering purposes, i.e., design of defect-tolerant structures, but scientific studies of materials often measure these properties as well. Laboratory fracture mechanics specimens impose severe mechanical conditions for fracture, and these conditions can promote fracture phenomena that are not revealed by other testing methods. For this reason, fracture mechanics-based materials tests are appealing for assessing hydrogen embrittlement. This section gives brief background information on fracture mechanics applied to structures and materials in hydrogen gas.

The average wall stress and the local stress near defects are related through the linear elastic stress-intensity factor (*K*). The magnitude of the local stress is proportional to the stress-intensity factor, *K*, according to the following relationship: [24,25]

$$\sigma_y = \frac{K}{\sqrt{2\pi x}} \tag{1}$$

where σ_y is the local tensile stress normal to the crack plane and x is distance in the crack plane ahead of the crack tip. The stress-intensity factor, *K*, is proportional to the wall stress and structural dimensions, *viz*:[24,25]

$$K = \beta \sigma_w \sqrt{\pi a} \tag{2}$$

where σ_w is the wall stress, the term β is a function of both defect geometry and structure geometry, and a is the defect depth.

Design parameters of structures containing defects can be established through the stress-intensity factor, K. The failure criterion for structures that contain defects and are subjected to static or monotonically increasing loads is as follows:

$$K \geq K_c \tag{3}$$

where K is the "applied" stress-intensity factor and K_c is the critical value of stress-intensity factor for propagation of the defect. The K_c value is a property of the structural material and can depend on variables such as the service environment. Combining Equations 2 and 3, the following relationship can be established:

$$\beta \sigma_w \sqrt{\pi a} \geq K_c \tag{4}$$

Equation 4 is the essential relationship for design of structures containing defects. Assuming K_c is known for the structural material and service environment, Equation 4 can be used in the following manner:

- If the structure dimensions and defect depth are known, the maximum wall stress can be calculated.
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- If the wall stress and defect depth are known, the structural dimensions can be calculated.

The failure criterion in Equation 4 pertains to structures subjected to static or monotonically increasing loads. Extension of a defect under these loading conditions is sustained as long as Equation 4 is satisfied. Defects can also extend by fatigue crack propagation when the structure is loaded under cyclic stresses. The rate of fatigue crack propagation is proportional to the stress-intensity factor range, i.e.:[24]

$$\frac{da}{dN} = C\Delta K^{n} \tag{5}$$

where da/dN is the increment of crack extension per load cycle, C and n are material- and environment-dependent parameters, and ΔK is the stress-intensity factor range. The stress-intensity factor range, ΔK, is defined as (K_{max} - K_{min}), where K_{max} and K_{min} are the maximum and minimum values of K, respectively, in the load cycle. The K_{max} and K_{min} are calculated from Equation 2. The relationship in Equation 5 is relevant for fatigue crack propagation at K_{max} values less than K_c but does not describe crack propagation in the lowest range of ΔK.

It must be noted that the fracture mechanics framework described above only applies when plastic deformation of the material is limited. Substantial plastic deformation may accompany propagation of existing defects in structures fabricated from relatively low-strength materials, e.g., carbon steels. In these cases, the linear elastic stress-intensity factor, K, does not accurately apply in structural design. Alternately, elastic-plastic fracture mechanics methods may apply.[24]

The hydrogen embrittlement susceptibility of structural steels can be quantified using fracture mechanics-based material properties. The critical values of stress-intensity factor for propagation of a defect under static and monotonically increasing loads in hydrogen gas are referred to as K_{TH} and K_{IH}, respectively,[7] in this chapter. For cyclic loading, the material response is given by the da/dN vs ΔK relationship measured in hydrogen gas. Enhanced hydrogen embrittlement is indicated by lower values of K_{TH} and K_{IH} but higher values of da/dN. Fracture mechanics properties of materials in hydrogen gas are typically measured under controlled laboratory conditions using standardized testing techniques.[26-28] These properties provide consistent, conservative indices of hydrogen embrittlement susceptibility.

4. Vessels and Pipelines in Hydrogen Energy Applications

An open question is whether steels currently used in hydrogen gas vessels and pipelines can be employed for similar applications in the hydrogen energy infrastructure. The answer to that question depends on several factors, including structural design constraints as well as steel properties. The information in section 2 demonstrates that steels are suitable structural materials provided hydrogen gas vessels and pipelines are operated within certain limits. In the proposed hydrogen energy infrastructure, it is anticipated that hydrogen gas vessels and pipelines will be subjected to service conditions that are outside the windows of experience. For example, hydrogen gas will likely be stored and transported at pressures that exceed those in current industrial gas and petroleum industry applications. The objective of this section is to provide insight into possible limitations on steel properties by illustrating trends in hydrogen embrittlement susceptibility as a function of important material, environmental, and mechanical variables.

The hydrogen embrittlement data in this section are for structural steels that are similar to those used in current hydrogen gas vessels and pipelines. In particular, data were selected for steels having compositions, microstructures, and tensile strengths that are germane to steels in hydrogen gas vessels and pipelines. In some cases, data are presented for steels having properties that deviate substantially from those used in gas vessels and pipelines. These cases are noted in the text, but the data trends still provide important insights. Fracture mechanics data were selected to demonstrate hydrogen embrittlement trends, since these data pertain to structures containing defects and provide conservative indices of fracture susceptibility in hydrogen gas.

Much of the data demonstrates that caution must be exercised in extending current steels to operating conditions outside the windows of experience. However, other data suggest that the hydrogen embrittlement resistance of steels can be improved.

4.1. Effect of Gas Pressure

Steels become more susceptible to hydrogen embrittlement as the materials are exposed to higher gas pressures. Thermodynamic equilibrium between hydrogen gas and dissolved atomic hydrogen is expressed by the general form of Sievert's Law:[17]

$$C = S\sqrt{f} \tag{6}$$

where *C* is the concentration of dissolved atomic hydrogen, the fugacity, *f*, of the hydrogen gas is related to the pressure (and temperature) of the system, and the solubility, *S*, of atomic hydrogen in the steel is a temperature-dependent material property. Equation 6 shows that as fugacity (pressure) increases, the quantity of atomic hydrogen dissolved in the steel increases; consequently, embrittlement becomes more severe. This trend is illustrated from K_{TH}, K_{IH}, and *da/dN* data. Figure 1 shows data for both low-alloy steels (K_{TH}) and carbon steels (K_{IH}), where critical *K* values decrease as hydrogen gas pressure increases for both types of steel.[10,29] Data for a low-alloy steel in Figure 2 demonstrate that *da/dN* measured at fixed stress-intensity factor range, ΔK, continuously increases as hydrogen gas pressure increases.[30] Finally, Figure 3 shows that increasing hydrogen gas pressure also accelerates *da/dN* in a carbon steel but only at lower ΔK values.[31]

The data in Figures 1 through 3 indicate that steel vessels and pipelines in hydrogen economy applications (i.e., at high hydrogen gas pressure) could be more vulnerable to hydrogen embrittlement than estimated from current experience. The quantities of hydrogen needed for a hydrogen-based economy suggest that gas could be stored and transported at pressures that exceed current limits. The American Society of Mechanical Engineers (ASME) is developing standards for hydrogen gas vessels with working pressures up to 100 MPa.[32] Current hydrogen gas vessels, however, have maximum working pressures in the range 20 to 30 MPa.[4] Figures 1 and 2 demonstrate that vessels fabricated from low-alloy steels become increasingly more susceptible to hydrogen embrittlement as pressures increase above 30 MPa. Current hydrogen gas pipelines are operated at pressures up to 13 MPa.[6] Figures 1 and 3 indicate that enhanced hydrogen embrittlement susceptibility must be considered for pipelines operating above 13 MPa.

4.2. Effect of Gas Impurities

Hydrogen gas embrittlement in steels can be altered by the presence of low concentrations of other gases in the environment. Certain gases such as oxygen can impede the adsorption of hydrogen gas on steel surfaces. Consequently, the kinetics of atomic hydrogen dissolution in steel can be greatly reduced, and the apparent hydrogen embrittlement determined from short-term testing is mitigated.[2,3] Sulfur-bearing gases such as hydrogen sulfide can have the opposite effect: the presence of these gases exacerbates hydrogen embrittlement.[33,34]

The effect of various gas additives on hydrogen embrittlement in a low-alloy steel is illustrated in Figure 4.[35] The data in Figure 4 show the ratio of fatigue crack propagation rate in hydrogen gas containing additives to fatigue crack propagation rate in hydrogen gas only. A ratio near 1.0 indicates that fatigue crack growth rates are equal in the two environments. The data demonstrate that oxygen and carbon monoxide gases in low concentrations can mitigate hydrogen embrittlement, while gases such as methyl mercaptan and hydrogen sulfide can compound hydrogen embrittlement.

The data in Figure 4 are effective in demonstrating the potential impact of a wide range of gas additives on hydrogen embrittlement for a single steel, however some further comments are needed. The low-alloy steel represented in Figure 4 was not heat treated by quenching and tempering, however the data trends are expected to apply to steel hydrogen vessels. Additionally, some studies confirm results from Figure 4, e.g., effects of oxygen and hydrogen sulfide,[33,34,36,37] however other studies report conflicting results. For example, Figure 4 shows that sulfur dioxide has no effect on fatigue crack propagation in hydrogen gas, but other studies have found that this gas species inhibits hydrogen embrittlement.[38] Finally, the measurements represented in Figure 4 were conducted for specific gas concentrations at a high load cycle frequency (i.e., 5 Hz), but such variables impact how severely gas additives affect hydrogen embrittlement.[39] Despite these caveats, the data in Figure 4 highlight the importance of trace gas constituents on environmental effects for steels in hydrogen gas.

The presence of non-intentional gas additives must be considered for hydrogen embrittlement of vessels and pipelines in the hydrogen energy infrastructure. The effect of gas impurities on hydrogen embrittlement may depend on the absolute partial pressure of the trace gas.[39] Increasing the operating pressure of vessels and pipelines will elevate partial pressures of impurities in hydrogen gas and potentially their role in hydrogen embrittlement.

Caution must be exercised in trying to exploit gas additives to control hydrogen embrittlement. While the data in Figure 4 suggest that gas additives such as oxygen could be employed to mitigate hydrogen embrittlement, the mechanistic role of gas additives must be considered. For example, oxygen is reported to impede the kinetics of atomic hydrogen uptake in metals such as steels, but over long time periods steels may dissolve sufficient hydrogen to suffer embrittlement. Therefore, gas additives that affect hydrogen uptake kinetics may impact manifestations of hydrogen embrittlement that operate at short time scales (e.g., fatigue loading) but not longer time scales (e.g., static loading).

4.3. Effect of Steel Strength

Hydrogen embrittlement in steels generally becomes more severe as material strength increases. This behavior arises because the magnitude of stress amplification near defects is proportional to material strength. These high stresses combined with the resulting enhanced hydrogen dissolution increase susceptibility to hydrogen embrittlement. The impact of material strength on hydrogen embrittlement is exemplified by the K_{TH} data in Figure 5.[10] Values of K_{TH} measured for low-alloy steels in hydrogen gas decrease as tensile strength, σ_{uts}, increases. A similar trend is expected for carbon steels.

Numerous studies have reported hydrogen embrittlement data trends similar to those in Figure 5.[40-43] However, some exceptions have been found in the literature. An example is provided in Figure 6, which shows fatigue crack propagation rate, *da/dN*, *vs* stress-intensity factor range, ΔK, plots for two low-alloy steels exposed to low-pressure hydrogen gas.[44] Crack propagation rates for the lower-strength steel (HY-80) exceed those in the higher-strength steel (HY-100) during exposure to hydrogen gas. The reason for the inconsistent hydrogen embrittlement trends portrayed in Figures 5 and 6 has not been determined, however it is important to note that data in the two figures were generated under two different loading formats. The K_{TH} data reflect crack growth under static loading, while the *da/dN* data pertain to fatigue crack growth under cyclic loading. Hydrogen-assisted crack growth under static loading is likely governed by crack tip stress, but hydrogen-assisted fatigue crack growth involves cyclic plastic strain. Crack propagation under these two modes of loading could be influenced by material strength differently. Additionally, fatigue crack growth rates can depend on the path of cracking through the steel microstructure. The difference in crack growth rates for HY-80 and HY-130 steels in Figure 6 could reflect effects of crack path and not solely material strength. The data in Figure 6 represent tests conducted in low-pressure hydrogen gas, but similar behavior is expected at higher gas pressure.

The effect of tensile strength on hydrogen embrittlement is important for vessels and pipelines in the hydrogen energy infrastructure, where high-strength materials may be attractive. Increasing the operating pressures of hydrogen gas vessels and pipelines could motivate the use of higher-strength steels. With increased gas pressure, the wall thickness of gas vessels and pipelines must increase to meet design stress requirements. However, with higher-strength steels, thinner walls can be used while maintaining the design stress. The data in Figure 5 demonstrate that steel vessels with tensile strength exceeding the current limits, i.e., 950 MPa,[4] will be more susceptible to hydrogen embrittlement under static loading. The data in Figure 6 suggest that higher-strength steels may be less susceptible to hydrogen-assisted fatigue crack growth.

4.4. Effect of Steel Composition

The concentrations of common elements in steels can significantly impact hydrogen embrittlement susceptibility. A striking demonstration of the effects of manganese, silicon, phosphorus, and sulfur on hydrogen embrittlement in a low-alloy steel is given by the data in Figure 7.[43] Values of K_{TH} are plotted *vs* the sum of bulk manganese, silicon, sulfur, and phosphorus concentrations. Examination of the steel compositions associated with individual data points in Figure 7 reveals that increases in manganese and silicon are detrimental to hydrogen embrittlement resistance, but variations in phosphorus and sulfur have little effect.

Similar trends were revealed from a study that individually varied elements such as manganese, sulfur, and phosphorus in a low-alloy steel.[40] Figure 8 shows that K_{TH} decreases as manganese increases from 0.07 to 2.65 wt%. Systematic variations in sulfur and phosphorus concentrations in the range 0.002 to 0.027 wt% did not affect K_{TH}. While the data indicate that variations in bulk sulfur and phosphorus in the concentration ranges examined do not alter the degree of hydrogen embrittlement, the presence of these elements is integral to the hydrogen embrittlement mechanism in low-alloy steels. While bulk compositions of sulfur and phosphorus should be minimized, the data show that additional benefit could be obtained by minimizing silicon and manganese as well. Although the low-alloy steels from Refs. [40] and [43] had extremely high strengths and were tested in low-pressure hydrogen gas, the trends in Figures 7 and 8 are expected to apply to lower-strength steels in high-pressure hydrogen gas.

The data in Figures 7 and 8 apply to low-alloy steels and may not give accurate insight into behavior for carbon steels. Increasing concentrations of manganese and silicon in low-alloy steels enhance the propensity for hydrogen-assisted fracture along grain boundaries.[43] Carbon steel fracture mechanics specimens tested under rising load in hydrogen gas do not exhibit fracture along grain boundaries, rather cracks propagate across the grains.[29] Since the role of manganese and silicon reflected in Figures 7 and 8 is to affect fracture along grain boundaries, the data trends probably do not describe behavior in carbon steels. Data showing effects of steel composition on K_{TH} or K_{IH} measured in hydrogen gas have not been found in the literature for carbon steels.

The hydrogen embrittlement resistance of low-alloy steels used in hydrogen gas vessels cannot be substantially altered by varying concentrations of elements such as manganese and silicon within the allowable composition ranges. Table 1 shows that the allowable composition ranges for manganese and silicon in 34CrMo4 steel are 0.50 to 0.80 wt% and 0.15 to 0.35 wt%, respectively. The data in Figure 7 indicate that K_{TH} noticeably improves only for manganese and silicon levels well below the lower limits in the 34CrMo4 steel composition ranges.

Altering composition may be one avenue to improve the hydrogen embrittlement resistance of steels. Vessels and pipelines in the hydrogen energy infrastructure will likely be subjected to higher gas pressures and may need to be fabricated from higher strength steels. Increasing either hydrogen gas pressure or steel strength will degrade resistance to hydrogen embrittlement. However, manufacturing steels with much lower manganese and silicon concentrations may balance the loss in hydrogen embrittlement resistance associated with increasing gas pressure or steel strength. Other data suggest that alloying elements not typically in the specifications for low-alloy steels could improve hydrogen embrittlement resistance. For example, data in Figure 8 show that additions of cobalt to a low-alloy steel with high tensile strength significantly increase K_{TH} values measured in low-pressure hydrogen gas.

4.5 Effect of Welds

Welding carbon and low-alloy steels can create residual stress and cause undesirable microstructure changes, e.g., formation of martensite, both of which make steel more vulnerable to hydrogen embrittlement.[9,21,23] Both the fusion zone and heat-affected zone regions of the weld can have microstructures that vary from the base metal.

Limited data show that both welding practice and location of defects can dictate the hydrogen embrittlement susceptibility of a weld. A study on microalloyed steel API 5L Grade X60 examined weld joints that were fabricated using either one or two weld passes.[45] Fracture mechanics specimens were extracted from the base metal, fusion zone, and heat-affected zone and tested in 7 MPa hydrogen gas. Results showed that K_{IH} values measured in the weld fusion zones were similar to values in the base metal; i.e., K_{IH} was approximately 100 MPa$\sqrt{m}$ in each region. In contrast, the heat-affected zones were more susceptible to hydrogen embrittlement, and K_{IH} was difficult to measure. The heat-affected zone in the two-pass weld was most susceptible.

Vessels and pipelines in the hydrogen energy infrastructure will be fabricated similar to current structures, where vessels are seamless and pipelines can be fabricated with both longitudinal welds and girth welds. Variables such as hydrogen gas pressure affect welds in a similar fashion to base metals, so the effect of increased gas pressure must be considered for hydrogen embrittlement of welds. Perhaps most important is the possibility of using steels that are outside the window of experience for hydrogen gas pipelines. Although hydrogen embrittlement at welds in current hydrogen gas pipelines has not been reported, it is acknowledged that the strength and microstructure of welds must be controlled to avoid hydrogen embrittlement.[5] The effect of alloy composition and welding practice on weld properties must be understood for any new steels used for hydrogen gas pipelines.

4.6. Effect of Mechanical Loading

Hydrogen embrittlement in steels can be manifested under different modes of mechanical loading, i.e., static, monotonically increasing, or cyclic. The severity of hydrogen embrittlement can depend on the specific mode of loading, e.g., static *vs* monotonically increasing, as well as variations in one type of loading.

Carbon and low-alloys steels having relatively low tensile strengths resist hydrogen embrittlement under static loads, but these alloys are susceptible under monotonically increasing loads. The carbon steel A516 exhibits hydrogen embrittlement when tests are conducted in hydrogen gas under rising-displacement loading (Figure 1).[29] However, cracks do not propagate in A516 steel when fracture mechanics specimens are statically loaded at K = 82 MPa√m in 70 MPa hydrogen gas.[10]

Variations in the rate of monotonic loading as well as the frequency and mean load for cyclic loading affect hydrogen embrittlement. Slow loading rates enhance hydrogen embrittlement, as demonstrated in Figure 9 for a low-alloy steel.[33] These K_{IH} measurements are for a high-strength steel tested in low-pressure hydrogen gas, but similar trends are expected for low-strength steels in high-pressure gas. Figure 10 shows that low load cycling frequencies increase fatigue crack growth rates for a carbon steel tested in hydrogen gas.[31] A similar effect of load cycle frequency on fatigue crack growth rate was measured for a low-alloy steel in hydrogen gas.[35] Finally, Figure 11 shows that fatigue crack growth rates in hydrogen gas do not depend on load ratio (i.e., K_{min}/K_{max}) for values up to 0.4.[46] However, over this range of load ratios, the difference in crack growth rates measured in hydrogen gas *vs* nitrogen gas diminishes. Crack growth rates in hydrogen gas increase at higher load ratios in Figure 11 because K_{max} approaches K_{IH} for the steel. Fatigue crack growth rates in hydrogen gas were also found to be independent of load ratio for the carbon steel ASME SA 105.[31] Similar effects of load cycle frequency and mean load on fatigue crack growth rates in hydrogen gas are expected for low-alloy steels.

Hydrogen vessels and pipelines in current applications are subjected to a variety of loading modes during service, including static, monotonically increasing, and cyclic. Vessels and pipelines in the hydrogen energy infrastructure are expected to experience these same modes of loading. At issue is whether operating conditions needed to support the hydrogen economy will cause substantial changes in variables such as loading rate and frequency as well as mean loads. For example, the in-line compressors needed for pipelines in the hydrogen energy infrastructure could alter the frequency and amplitude of pressure fluctuations compared to current pipelines. In addition, hydrogen gas vessels could be filled and emptied more frequently in the hydrogen economy. The data in Figures 9 and 10 suggest that higher loading rates and frequencies mitigate hydrogen embrittlement in structural steels. However, actual duty cycles involve sequences of active and static loads that are more complex than the uniform loading conditions used in laboratory tests. Hydrogen embrittlement data generated under loading conditions that mimic real duty cycles are needed to better understand the impact of mechanical loading variables on hydrogen gas vessels and pipelines.

5. Conclusion

Experience with steel vessels and pipelines in the industrial gas and petroleum industries demonstrates that these structures can be operated safely with hydrogen gas, although the experience is limited to certain ranges of material, environmental, and mechanical variables. Gas pressures in vessels and pipelines for the hydrogen economy are certain to exceed the limit in current applications. Data consistently show that steels are more susceptible to hydrogen embrittlement at higher gas pressures. As operating pressures increase, designs will demand higher strength materials. Most data indicate that steels are more vulnerable to hydrogen embrittlement when strength increases. The effects of other variables, such as gas impurities, welds, and mechanical loading on hydrogen embrittlement of steel vessels and pipelines in the hydrogen economy are not as certain. Hydrogen embrittlement resistance of steels could be improved through production of low-manganese and low-silicon steels. Data for high-strength steels in low-pressure hydrogen gas show that composition has a dramatic effect on hydrogen embrittlement, however this trend has not been demonstrated for lower-strength steels in high-pressure hydrogen gas.

Although hydrogen embrittlement is more severe at high gas pressures and in high strength steels, structures can still be designed with steels under these conditions by using fracture mechanics. Provided material data is available for steels in high-pressure hydrogen gas, the limiting crack depth, wall stress, and structure dimensions can be defined using fracture mechanics.

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Table 1. Composition (wt%) of 34CrMo4 steel[a]

Cr	Mo	C	Mn	Si	P	S	Fe	P+S
0.90 1.20	0.15 0.25	0.30 0.37	0.50 0.80	0.15 0.35	0.035[b] max.	0.035[b] max.	balance	0.04[b] max.

[a]The composition limits for 34CrMo4 vary slightly among European countries. The specification in Table 1 is from Germany.[4] The 34CrMo4 steel composition is almost identical to either AISI 4130 or AISI 4135 steel.[47]
[b]Limits for P and S in new hydrogen gas vessels are 0.025 wt%.

Table 2. Composition (wt%) of A106 Grade B steel[a]

C	Mn	P	S	Si	Cr	Cu	Mo	Ni	V	Fe	Cr+Cu+Mo+ Ni+V
0.30 max.	0.29 1.06	0.035 max.	0.035 max.	0.10 max.	0.40 max.	0.40 max.	0.15 max.	0.40 max.	0.08 max.	balance	1.0 max.

[a]Specification is for seamless pipe.[48]

Table 3. Composition (wt%) of API 5L steels[a]

	C	Mn	P	S	Nb+V+Ti	Fe
Grade X42	0.22 max.	1.30 max.	0.025[b] max.	0.015[b] max.	0.15 max.	balance
Grade X52	0.22 max.	1.40 max.	0.025[b] max.	0.015[b] max.	0.15 max.	balance

[a]Product Specification Level 2 composition for welded pipe.[49]
[b]Recommended maximum concentrations of P and S are 0.015 and 0.01 wt%, respectively, for modern steels in hydrogen gas service.[5]

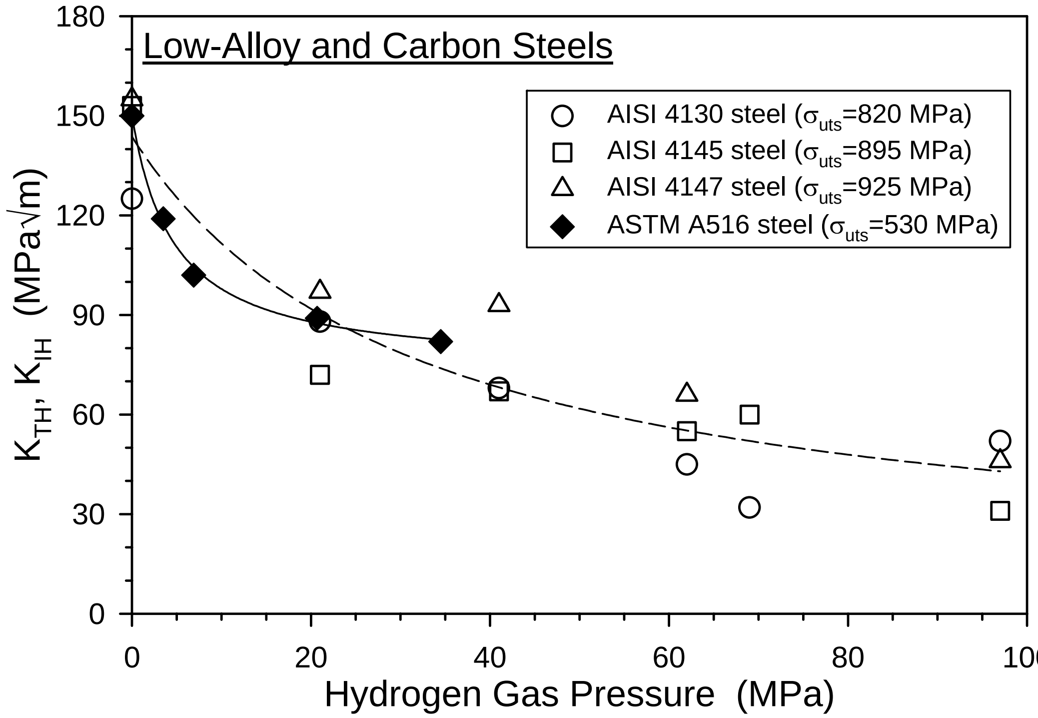

Figure 1. Effect of gas pressure on critical stress-intensity factor for crack extension in hydrogen gas (K_{TH} or K_{IH}).[10,29] The low-alloys steels (open symbols) were tested under static loading, while the carbon steel (filled symbols) was tested under rising displacement loading. Data points at zero pressure represent fracture toughness measurements in air, i.e., K_{Ic}.

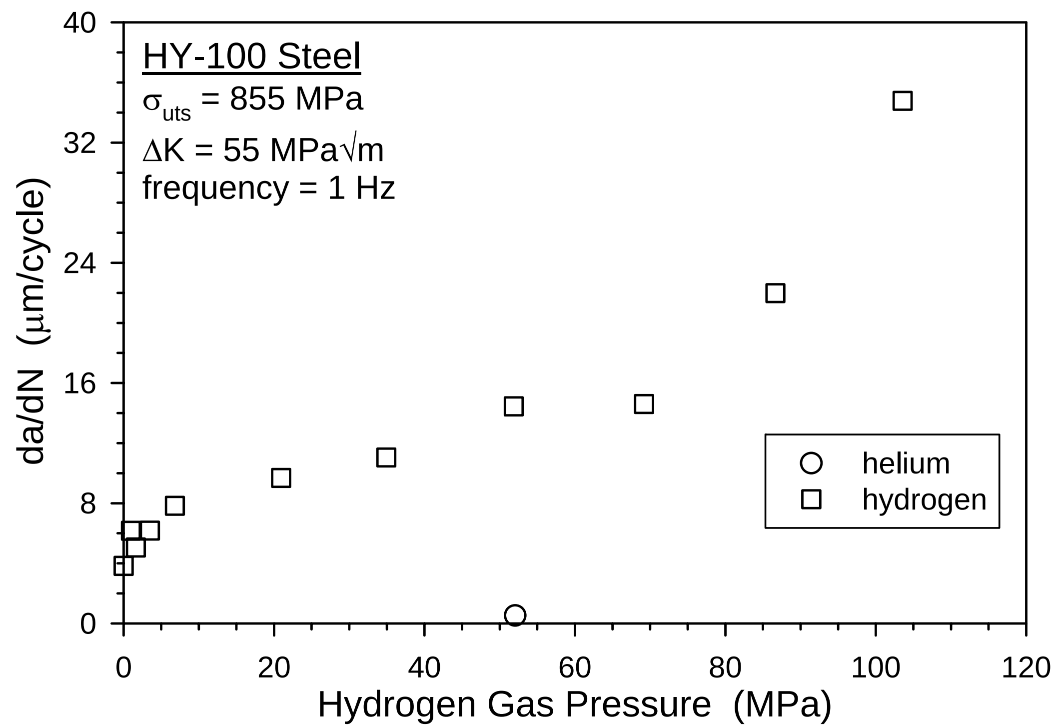

Figure 2. Effect of hydrogen gas pressure on fatigue crack growth rate (*da/dN*) at constant stress-intensity factor range (ΔK) in a low-alloy steel.[30]

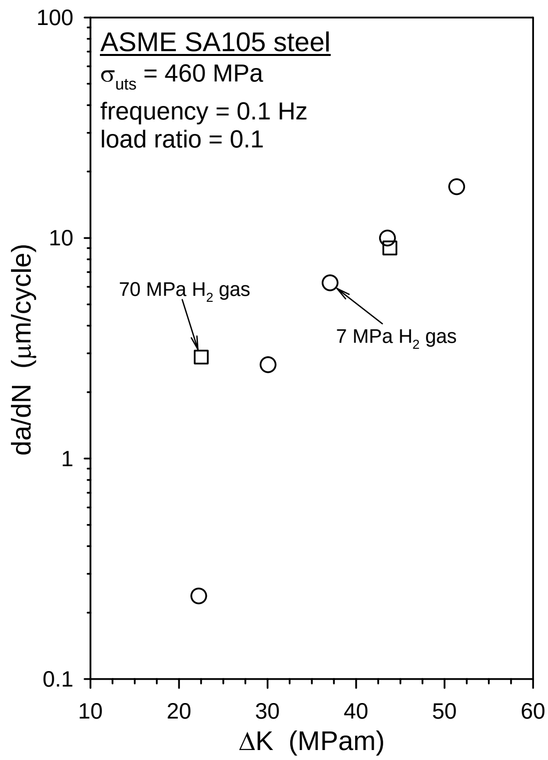

Figure 3. Effect of hydrogen gas pressure on fatigue crack growth rate (*da/dN*) *vs* stress intensity factor range (*ΔK*) relationships for a carbon steel.[31]

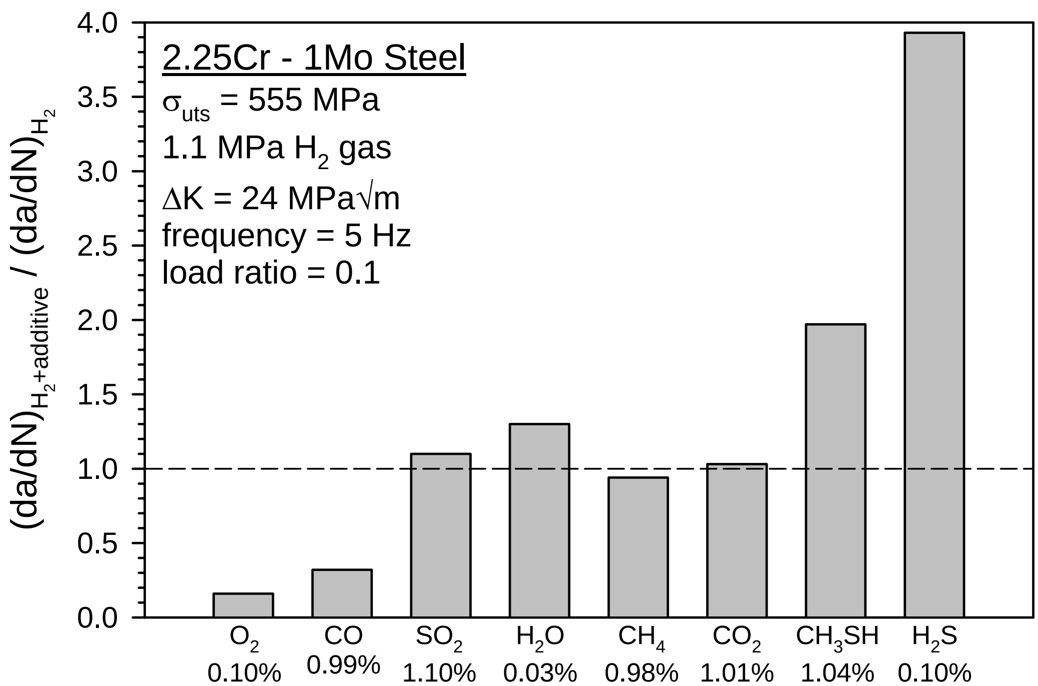

Figure 4. Effect of gas additives on the fatigue crack growth rate (*da/dN*) at constant stress-intensity factor range (*ΔK*) for a low-alloy steel in hydrogen gas.[35]

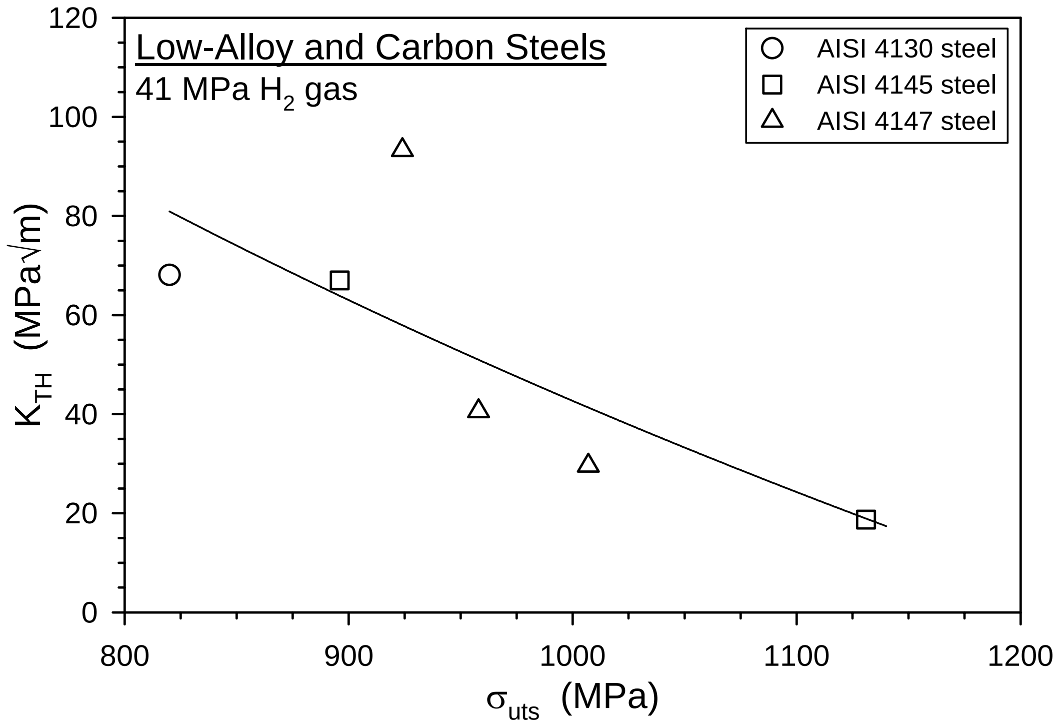

Figure 5. Effect of tensile strength (σ_{uts}) on critical stress-intensity factor for crack extension in hydrogen gas (K_{TH}).[10] Data is for low-alloy steels tested under static loading.

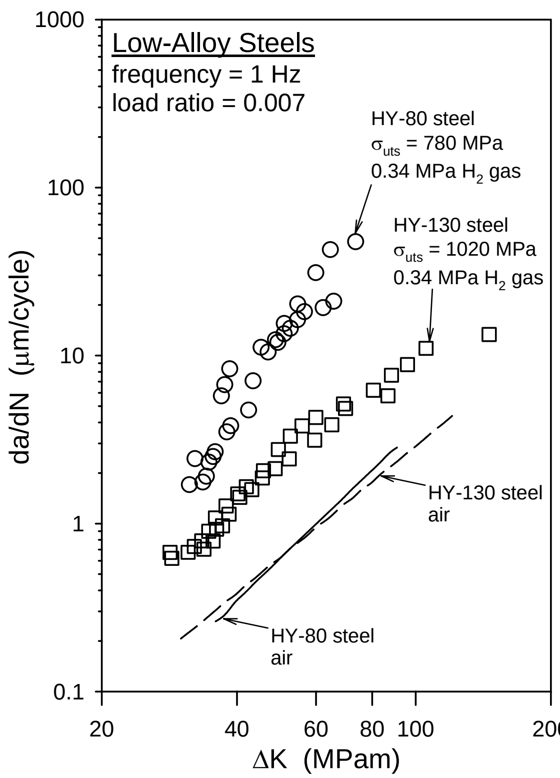

Figure 6. Fatigue crack propagation rate (*da/dN*) *vs* stress-intensity factor range (ΔK) relationships measured in low-pressure hydrogen gas for two low-alloy steels with different tensile strengths.[44]

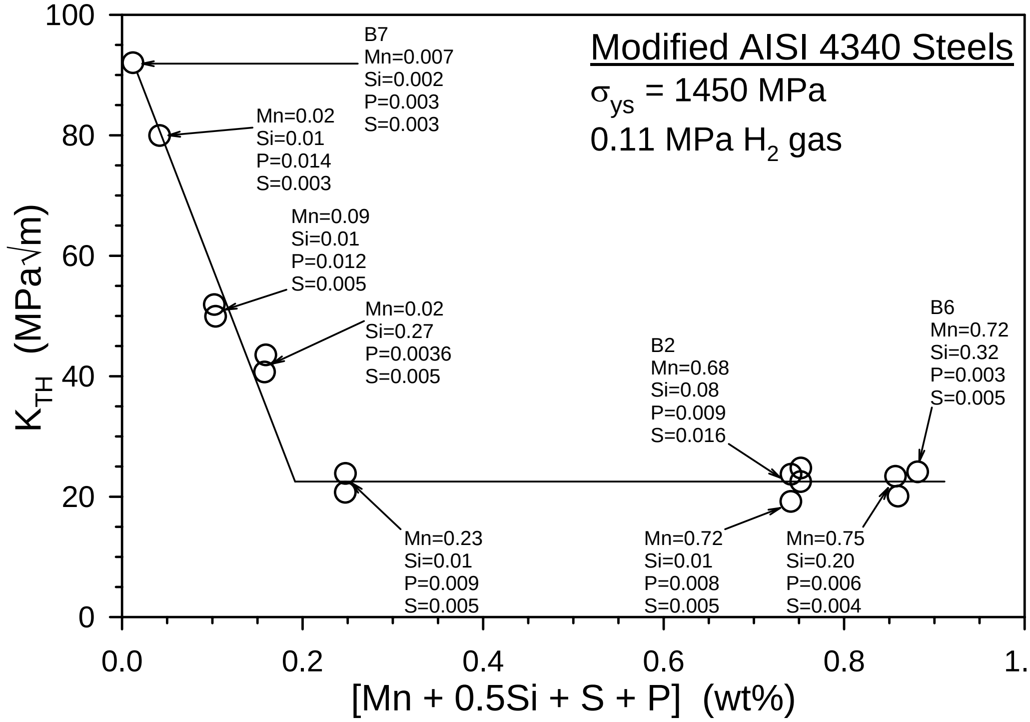

Figure 7. Effect of manganese, silicon, phosphorus, and sulfur content on critical stress-intensity factor for crack extension (K_{TH}) in low-alloy steels.[43] Data is for high-strength steel tested in low-pressure hydrogen gas.

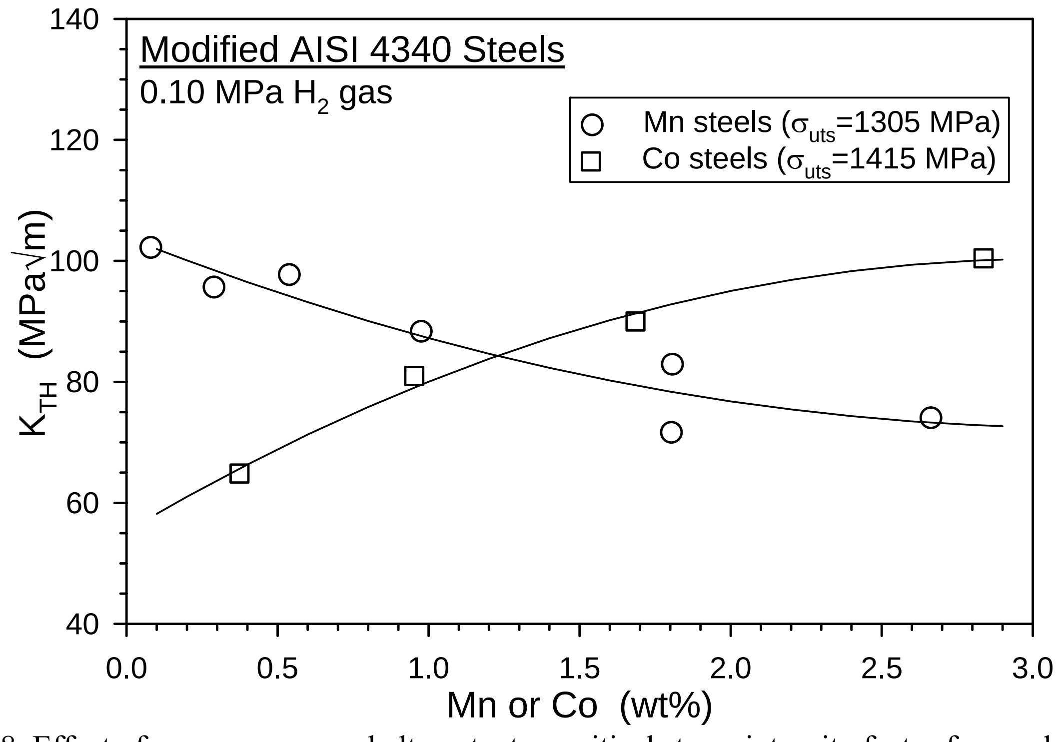

Figure 8. Effect of manganese or cobalt content on critical stress-intensity factor for crack extension (K_{TH}) in low-alloy steels.[40] Data is for high-strength steel tested in low-pressure hydrogen gas.

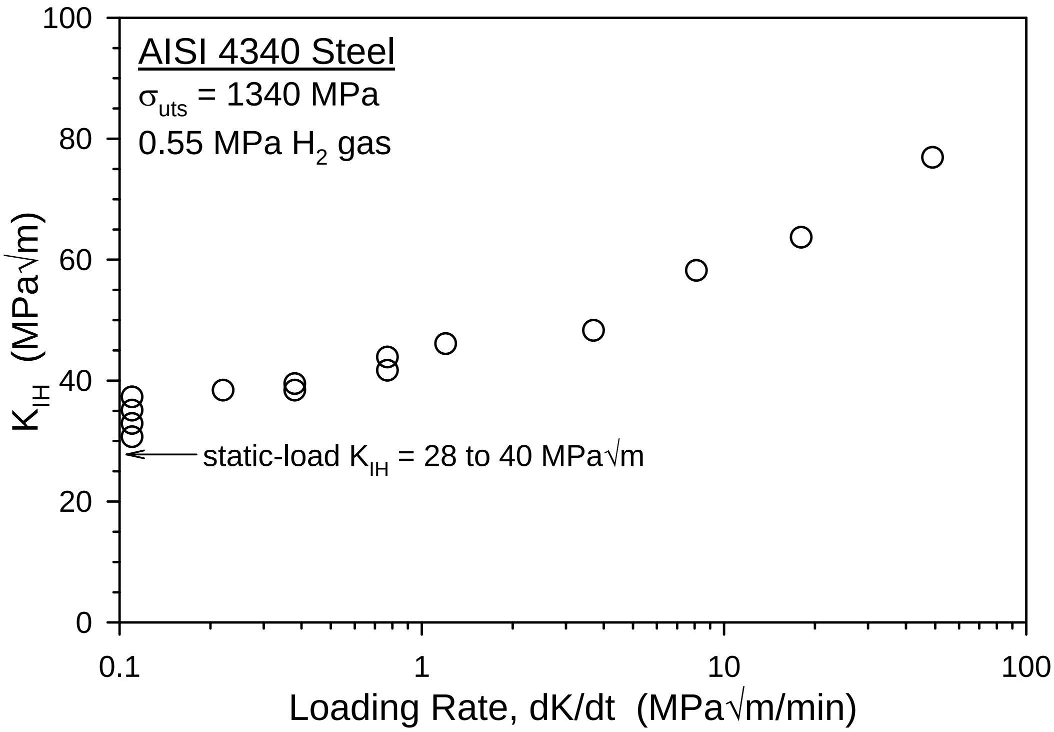

Figure 9. Effect of loading rate (*dK/dt*) on critical stress-intensity factor for crack extension (K_{IH}) in a low-alloy steel.[33] Data is for high-strength steel tested in low-pressure hydrogen gas.

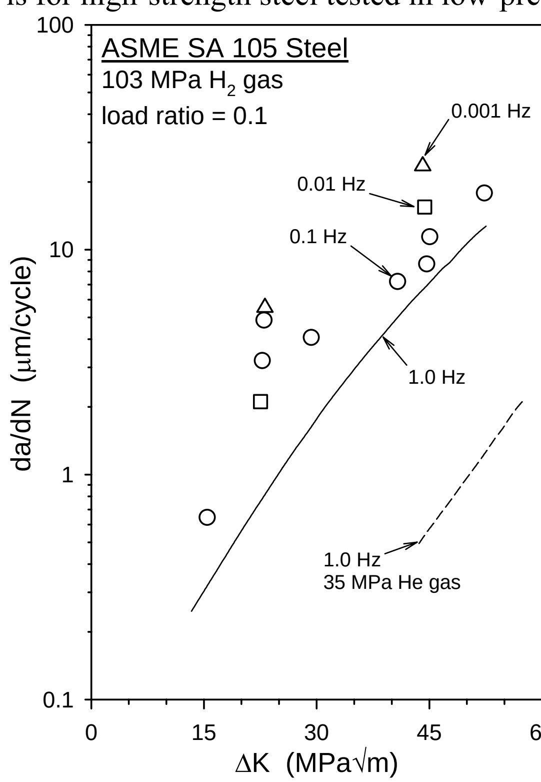

Figure 10. Effect of load cycle frequency on fatigue crack growth rate (*da/dN*) *vs* stress intensity factor range (ΔK) relationships for a carbon steel.[31]

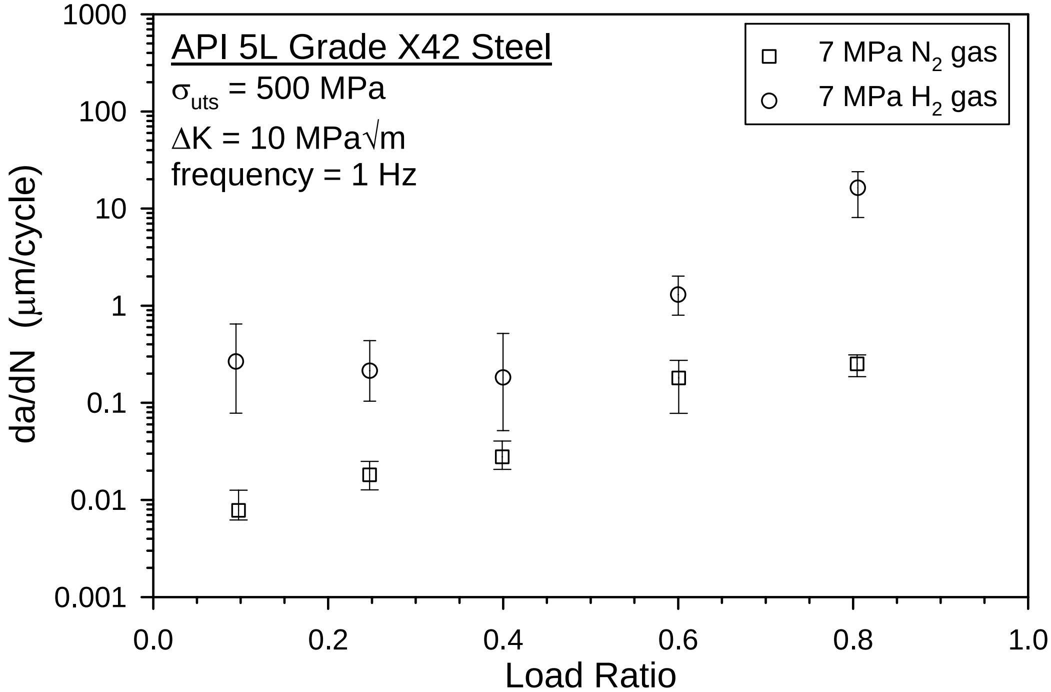

Figure 11. Effect of load ratio (ratio of minimum load to maximum load) on fatigue crack growth rate (*da/dN*) at fixed stress intensity factor range (ΔK) in hydrogen gas for a carbon steel.[46]