

Potential Effects of Hydrogen on Materials in Appliances

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Table of contents

04 Executive summary

05 Background and motivation

06 Effects of hydrogen on metals

10 Effects of hydrogen on polymers

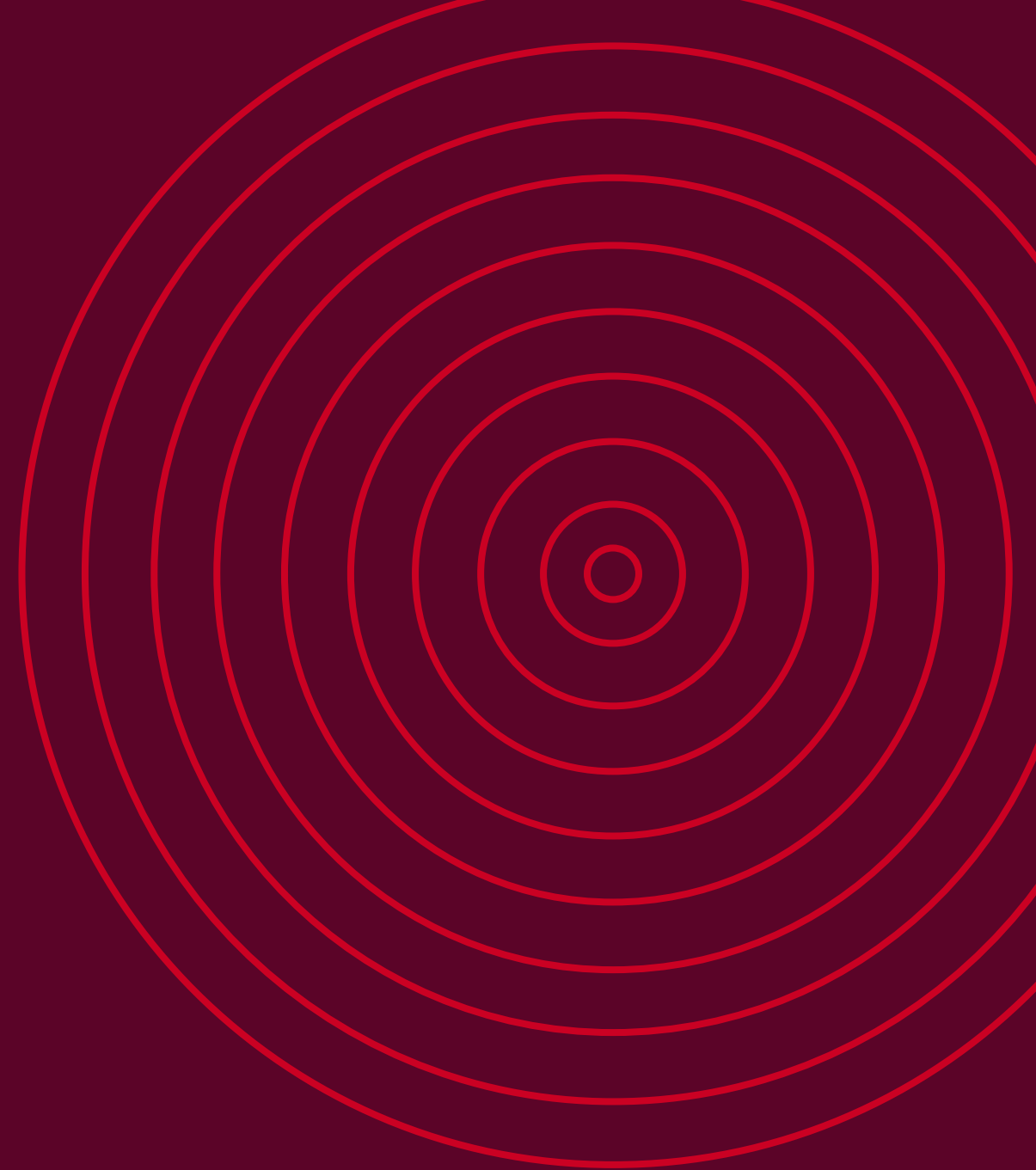
13 Summary

14 Endnotes

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Executive summary

With the global focus on moving to renewable energy sources, there is an increased interest in reducing the use of fossil fuels, including natural gas. As part of this effort, hydrogen gas is being considered either as a substitute for natural gas or to be used as a blend with natural gas. Hydrogen is of interest since there are methods to produce it using renewable energy sources (generating what is known as green hydrogen) and burning hydrogen instead of natural gas may also reduce pollutants. This literature review looks at the compatibility of hydrogen and hydrogen-natural gas blends with some materials that may be found in current natural gas-fired appliances, such as ranges, boilers, and water heaters.

The potential for hydrogen embrittlement in several metals that may be used in gas-fired appliances, namely plain carbon steels, cast irons, stainless steels, copper alloys, and aluminum and its alloys, was investigated. Also considered were the stability and permeability of polymers in their potential uses in gas-fired appliances. Since hydrogen gas molecules are smaller than the methane molecules primarily making up natural gas, knowing the ability of these polymers to contain hydrogen is critical. Some applications of polymers include their use in seals and valves, and leaking could lead to fire and explosion hazards.

Potential effects of hydrogen on materials in appliances

Background and motivation



Currently, the world is looking for solutions to achieve carbon neutrality and is facing an energy shortage, especially in Europe. Many countries are focused on “green” hydrogen produced by renewable energy because it could play a key role in helping the world achieve a greenhouse gas-neutral economy by 2050. The addition of hydrogen to natural gas is being considered as an efficient means to utilize existing infrastructure while decreasing the carbon intensity of the energy supply. The European Commission (EC) expects 1.3 million tons of green hydrogen to be blended into the natural gas network by 2030.¹ The permissible volume percentage of hydrogen in natural gas networks varies globally with regional regulation and ranges from 0% up to 20%. Additionally, there is discrepancy on what hydrogen concentrations current gas-fired appliances would need no or only minor modifications to safely function.²

An important concern is the effect of hydrogen on materials. The natural gas infrastructure, including appliances, contains several different metals and polymers. It is known that hydrogen can affect certain properties of metals in different ways. Polymers also make up a large proportion of the materials used in natural gas appliances, in particular the seals used at gas connections and in gas valves. Of particular concern for seals is leakage since hydrogen is the smallest and lightest molecule. Since it is so much smaller than a methane molecule, hydrogen could leak through an existing seal. In addition to leaking, hydrogen has been known to change the permeability and stability of polymers. This report looks at current literature to determine whether hydrogen would have a negative effect on the properties of materials that are likely to be found in appliances.

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Effects of hydrogen on metals

When exposing steel to hydrogen, it should be considered whether hydrogen embrittlement will be an issue. Hydrogen embrittlement (HE) manifests itself in multiple ways, including failures due to cracking, blistering, hydride formation, and a reduction in the tensile ductility of the metal. These failures occur at tensile stresses (including residual stresses) below the rated strength of the steel.³ Hydrogen-induced cracking, which is a type of hydrogen embrittlement, manifests itself as a time-delayed fracture at a point of tension even though the part is loaded well below its tensile strength. This is due to stepwise internal cracking connecting hydrogen blisters within the metal. Higher-strength steels are particularly susceptible to this type of hydrogen embrittlement resulting in catastrophic brittle failure.⁴ Generally, it is accepted that this includes steels with a tensile strength greater than 1,000 MPa (140 ksi) or with a hardness greater than 30 HRC (301 HV).⁵ ASME B31.12 uses more conservative limits, setting a maximum hardness limit of 21 HRC (235 HV) for use with hydrogen piping and pipelines, which correlates to an estimated tensile strength of 783 MPa (114 ksi).⁶ That is not to say that lower strength steels, which are more likely to be used in appliances, will not be affected by

the exposure to hydrogen. It is likely that such steels will lose some of their tensile ductility, but research [Zhou Z.] indicates that their strength will not be affected to the point where they will fail far below their rated strengths.³

Hydrogen also accelerates fatigue crack growth and reduces the fatigue endurance limits of all carbon and low alloy steels. This is due to hydrogen reducing the ductility of steel. This would be a concern if there are expected to be fluctuations in the gas pressure. Also because of this deleterious effect on steel's ductility, the crack growth rate from existing defects may be accelerated.³ The percentage of hydrogen in a hydrogen-natural gas blend may also play a part in determining to what degree, if any, steels will be affected. It has been found [Ronevich] that accelerated fatigue crack growth was notable at 5% hydrogen, with increased hydrogen percentages contributing little additional acceleration.⁷ Considering the low gas pressure in the appliances (and, consequently, low magnitude of expected pressure fluctuations) and the stresses being such a low percentage of the strength of typical steels in general, it has been reported [Zhou] that hydrogen-enhanced crack growth may not be a concern.³

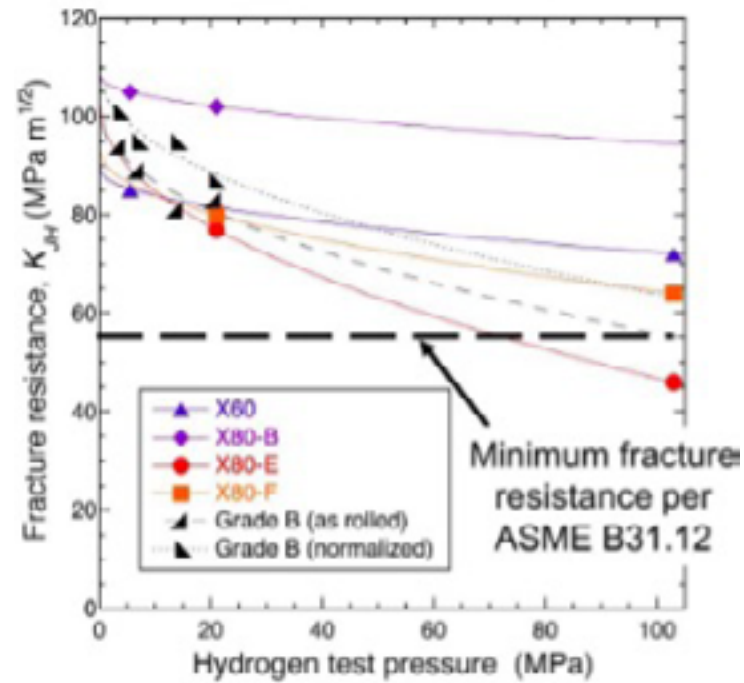


Figure 1: Fracture resistance of X60 and X80 steels with increased hydrogen (100%) test pressure.⁸

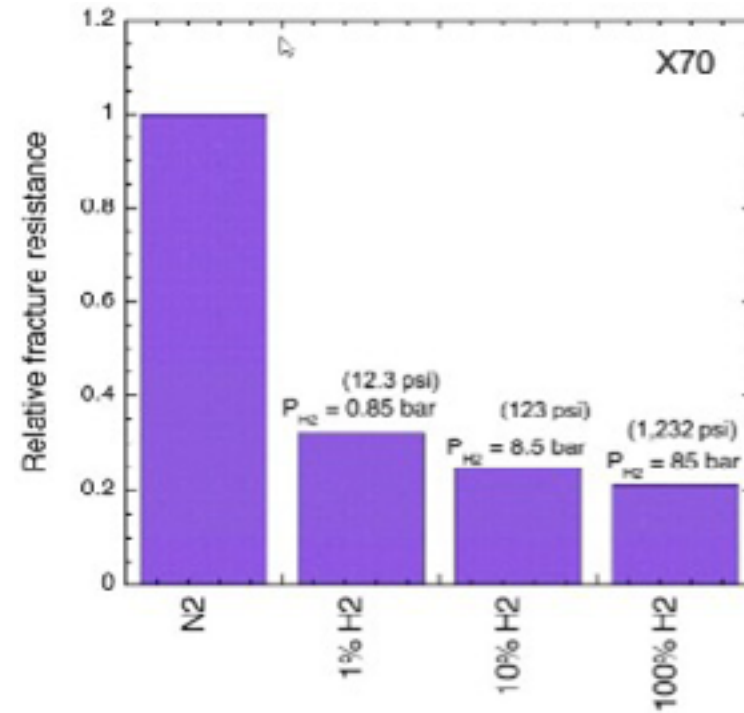


Figure 2: Illustration of how the relative fracture resistance of X70 steel is affected by hydrogen gas even at low pressures. This graph also shows that the effect from 100% hydrogen gas is not much different than 1% hydrogen gas (nitrogen used as the balance gas during testing).⁷

Some data indicates that the effect of hydrogen gas on fracture toughness depends on the pressure of the gas, as is illustrated in Figure 1.⁸ The pressures in the pipes distributing natural gas to appliances are generally considered low and range from 0.25 psig to 60 psig (1.72 kPa-413 kPa), though sometimes the pressure can be as high as 100 psig (690 kPa).³ However, even at relatively low pressures, hydrogen gas does cause a notable decrease in fracture toughness and this decrease in fracture toughness is not much different for a 1% blend of hydrogen compared to 100% hydrogen, as is illustrated in Figure 2.⁷ This figure shows that the effect on fracture toughness can be substantial at any percentage of hydrogen and at any pressure level.

It can be valuable to know whether hydrogen embrittlement is a problem for the other metals that may be present in natural gas appliances, which can include cast iron, stainless steel, copper, brass, and aluminum. Though perhaps not pertinent to appliances, it is also known that nickel, titanium, and their alloys are known to be prone to hydrogen embrittlement and are not generally considered for hydrogen service.⁹

As with steel, cast iron is also affected by hydrogen embrittlement. Due to the general mechanical properties of cast irons being of lower strength and hardness than most steels, the concerns of hydrogen embrittlement are essentially the same as those of lower strength steels, namely that hydrogen can decrease the ductility of cast iron, increase its potential crack growth rate, and decrease

its fracture toughness. Due to the already somewhat brittle nature of cast iron without hydrogen exposure, further embrittlement due to hydrogen may limit its usability. That said, some are considering ductile iron (a category of cast iron) for equipment exposed to hydrogen due to its potentially favorable cost, workability, and formability.¹⁰ However, the usability of cast iron will depend on relevant necessary mechanical properties and how hydrogen exposure affects the cast iron (which may be related to cast iron composition and processing treatments). Several teams are investigating how hydrogen embrittlement affects different cast irons.^{11,12,13} At least one organization [GPA Engineering] claims that hydrogen embrittlement is a negligible concern for cast iron in appliances.¹⁴

Stainless steels are not considered any more prone to hydrogen embrittlement than plain carbon types of steel. That said, austenitic stainless steels are known to be less susceptible to hydrogen embrittlement than ferritic stainless steels. In fact, 304L and 316L stainless steels are currently used in hydrogen gas service.¹⁵ For ferritic stainless steels, their resistance to hydrogen embrittlement is reduced with increased heat treatment temperature and molybdenum content as well as applying stresses perpendicular to the cold-rolling/cold-working direction of the metal. Austenitic stainless steels in the annealed or only lightly cold-worked condition can be very resistant to hydrogen cracking. However, their resistance decreases if they are highly cold-worked. This is attributed to increased yield strength and the formation of martensite when the material is highly cold-worked. For austenitic stainless steels with a highly stable austenite phase and high yield strength (for example, 21Cr-6Ni-9Mn), their susceptibility is deemed solely dependent on their yield strength, which is similar to the behavior of low-alloy steels.¹⁶

Considering high-strength stainless steels in a sulfide-free environment, it was found that martensitic and precipitation-hardened stainless steels can be prone to

hydrogen embrittlement at yield strengths above 1035 MPa (though this is for smooth specimens; for notched or pre-cracked specimens, hydrogen embrittlement may occur at lower yield strengths). Tempering or over-aging can increase the hydrogen embrittlement resistance of martensitic and precipitation-hardened stainless steels by effectively lowering their yield strengths. For environments that contain sulfides, it is known that the presence of hydrogen sulfide in the environment decreases the resistance to hydrogen embrittlement of high-strength stainless steels. Studies have shown that no stainless steel fully resists hydrogen embrittlement in sulfide-present environments if their yield strength is greater than 690 MPa.¹⁶

Copper and its alloys are generally not considered susceptible to hydrogen attack unless they contain cuprous oxide (Cu_2O), and since most copper alloys are deoxidized, they are consequently not prone to hydrogen embrittlement. When cuprous oxide-bearing copper is heated in a hydrogen or hydrogen-blend gas, the hydrogen diffuses into the copper to react with the oxide to form water. With additional heat (375 °C), this water becomes high-pressure steam that produces fissures, effectively decreasing the ductility of the metal (generally called hydrogen embrittlement). Consequently, tough pitch coppers, which contain small quantities of Cu_2O , are prone to some degree of hydrogen embrittlement. An example of a tough pitch copper is C11000. Deoxidized coppers that have low residual deoxidizer contents, such as C12000, can contain some low levels of Cu_2O and are not immune to hydrogen embrittlement. However, deoxidized coppers with high residual deoxidizer contents are not considered susceptible to hydrogen embrittlement because the oxygen present is combined into complex oxides that are not prone to reacting with hydrogen. The resistance of copper to hydrogen embrittlement (except in the cases discussed above) also includes copper alloys.

These alloys include: low-zinc brasses, high-zinc brasses, special brasses, phosphor bronzes, copper nickels, nickel silvers, tin bronzes, leaded tin bronzes, high-lead tin bronzes, leaded red brasses, leaded semi-red brass, leaded yellow brasses, leaded high-strength yellow brasses, high-strength yellow brasses, aluminum bronzes, leaded nickel brasses, leaded nickel bronzes, silicon bronzes, and silicon bronzes (as defined in the ASM Specialty Handbook: Copper and Copper Alloys).¹⁷ In a test looking at the potential compatibility of copper and its alloys in a hydrogen-natural gas blend system, copper and two types of brass (CW617N and CW614N) were tested with a 20% hydrogen blend and did not show significant susceptibility to hydrogen embrittlement.¹⁵ Despite the resistance of copper and copper alloys to hydrogen embrittlement, it has been reported that soldered or brazed copper joints may not be compatible in systems utilizing hydrogen. This is due to a proposition that, if there were to be a leak (which is perhaps more likely due to the small molecular size of hydrogen gas) that ignited into a flame, the surrounding material should be able to withstand heat from the flame for a short period of time. The metals used for soldering and brazing copper joints have lower melting temperatures than the surrounding materials and may not be able to withstand heat from such a flame as robustly as other joint types.⁹ Note that copper is not permitted for use without a coating per gas appliance standards; however, there is not a current requirement that the coating prevent hydrogen from reaching the copper base material.

It has been claimed that aluminum is not susceptible to hydrogen embrittlement when exposed to dry hydrogen gas due to difficulty in hydrogen permeating the aluminum oxide formed at its surface as well as low hydrogen solubility and diffusivity in aluminum.^{15,18} This dry hydrogen gas condition is seemingly what will be encountered by appliances typically used with natural gas. That said, in aqueous environments, such as in the presence of water vapor or any gas containing water vapor, it is known that aluminum and its alloys are prone to hydrogen uptake and, consequently, hydrogen embrittlement.¹⁹ The extent of this embrittlement seems highly dependent on the grain structure of the material as well as the presence of hydrogen-trapping particles within the material. This is according to a report [Huan Zhao et al] that claims hydrogen only seems to embrittle the material at the grain boundaries of aluminum whereas particles in the bulk of the material act to trap the hydrogen and hinder crack propagation.²⁰ In addition, for an appliance that utilizes aluminum near a gas burner, any potential for elevated temperatures due to burning hydrogen may need to be assessed to ensure that melting of the aluminum would not be a problem.⁹

Effects of hydrogen on polymers

Polymers are not subject to hydrogen embrittlement in the same ways as metals. Hydrogen absorbed by polymers exists as a diatomic molecule, and it does not dissociate as it is known to do in metals.

The properties of polymers depend not only on their chemical structure, such as chain length, side groups, branching and crosslinking, but also on several other important factors. One is the molecular weight of polymer chains, and the other is processing history. Furthermore, fillers, plasticizers, crosslinking agents, flame retardants, etc., are often incorporated to modify the properties of polymers. The cooling rate of the molten state also changes the degree of crystallinity of polymers. Some standards and chemical compatibility handbooks show that some polymers are stable in a gaseous hydrogen atmosphere. **Table 1** illustrates some common polymers considered compatible with gaseous hydrogen as well as the sources citing their compatibility.

Chemical name	Trade name	Source
Polytetrafluoroethylene (PTFE)	Teflon	ISO 15916 ²¹
Polychloroprene (CR)	Neoprene	ISO 15916
Polyester fiber	Dacron	ISO 15916
Polyester film	Mylar	ISO 15916
Nitrile	Buna-N	ISO 15916
Polyamide (PA)	Nylon	ISO 15916
Polychlorotrifluoroethylene (PCTFE)	Kel-F	ISO 15916
Nitrile rubber (NBR)	Buna N	Chemical compatibility (Emerson) ²²
Fluoroelastomers of vinylidene fluoride (FKM)	Viton	Chemical compatibility (Emerson)
Ethylene propylene (EPDM)		Chemical compatibility (Emerson)
Perfluoroelastomer (FFKM)		Chemical compatibility (Emerson)
Thermoplastic elastomer (TPE)		Chemical compatibility (Graco) ²³
Polypropylene (PP)		Chemical compatibility (Graco)
Polytetrafluoroethylene (PTFE)		Chemical compatibility (Graco)
Ethylene propylene rubber (EPR)		Chemical compatibility (Graco)

Table 1: Examples of polymers that are considered compatible with hydrogen and the corresponding sources where that compatibility is supported.

The seals used in gas connections and valves in some appliances are predominantly made of polymeric materials. Sealing materials are typically elastomeric materials such as nitrile rubber (NBR), fluoroelastomers of vinylidene fluoride (FKM), copolymer of ethylene and propylene (EPM), fluorosilicone (FMQ), silicone (MQ), polychloroprene (CR) etc., which have a relatively narrow temperature range for standard operation. Semicrystalline thermoplastics, such as polytetrafluoroethylene (PTFE), polyetheretherketone (PEEK), polyamide (PA), polyimide from pyromellitic dianhydride and 4,4' diamino diphenyl ether, polychlorotrifluoroethylene (PCTFE), etc., are also used in sealing applications and have the advantage that they can be used over a much wider range of temperatures.

Table 2 lists the common components of end-use appliances and examples of associated polymeric materials.²⁴ This table may not include all the polymers used in end-use appliances.

Table 2: Examples of polymers used in natural gas end-use appliances.²⁴

Component	Description	Polymers (example)
Flange connections	O-rings, gaskets	<ul style="list-style-type: none"> Nitrile rubber (NBR) Fluoroelastomers of vinylidene fluoride (FKM) Polytetrafluoroethylene (PTFE)
Valves	Pistons	Polyetheretherketone (PEEK)
Valves	O-rings, fittings	<ul style="list-style-type: none"> Nitrile rubber (NBR) Fluoroelastomers of vinylidene fluoride (FKM) Polytetrafluoroethylene (PTFE)
Valves	Seals and gaskets	<ul style="list-style-type: none"> Nitrile rubber (NBR) Fluoroelastomers of vinylidene fluoride (FKM) Polytetrafluoroethylene (PTFE) Polytetrafluoroethylene (EPM) Fluorosilicone (FMQ) Silicone (MQ) Polychloroprene (CR) Polyamide (PA) Polyetheretherketone (PEEK)
Valves	Valve seats	<ul style="list-style-type: none"> Polyamide (PA) Polyimide from pyromellitic dianhydride and 4,4' diamino diphenyl ether Polychlorotrifluoroethylene (PCTFE) Polytetrafluoroethylene (PTFE)

Polymer materials such as semicrystalline thermoplastic or elastomeric materials have free volume between molecular chains caused by the segmental motion of the molecules. Therefore, the dihydrogen could easily diffuse into a bulk material and permeate from the material. There are multiple methods to determine the permeability properties of a gas, such as the gravimetric method, the manometric method, the constant pressure method, the differential pressure method, the pressure sensor method and the thermal desorption analysis gas chromatography (TDA GC) method. To allow for the conformity assessment of permeability properties, some methods have been standardized, as illustrated in **Table 6**. Besides the seals at valves and other internal connections, residential gas hose connections to appliances need to be considered for leakage, due to hydrogen gas having a smaller molecular size than methane, and the resulting fire/explosion risk. These connection types have a substantial historical relation with residential explosions, particularly with older connectors.^{32,33,34}

Table 3: Examples of standardized methods for permeability properties.

Measurement method	ASTM	ISO
Polytetrafluoroethylene (PTFE)	Teflon	ISO 15916 ²¹
Polychloroprene (CR)	Neoprene	ISO 15916
Polyester fiber	Dacron	ISO 15916
Polyester film	Mylar	ISO 15916
Nitrile	Buna-N	ISO 15916
Polyamide (PA)	Nylon	ISO 15916

Summary

In determining whether appliances that are currently used with natural gas can be used with hydrogen gas or a hydrogen-natural gas blend, one important consideration is whether there are any potential incompatibility issues with the materials that are in the presence of hydrogen. Based on the literature reviewed, it was seen that hydrogen-induced cracking is of a particular concern for high-strength steels and that hydrogen embrittlement reduces ductility, accelerates fatigue crack growth and reduces fatigue endurance limits, and reduces the toughness for all carbon and low alloy steels. Cast irons are affected in much the same way as steels, with the hydrogen-induced reduction in ductility of these materials a notable concern since these materials are already somewhat brittle to begin with. It was also seen that some stainless steels are resistant to hydrogen attack (for example, certain austenitic stainless steels), while other stainless steels may be more prone to hydrogen embrittlement. It was seen that copper and its alloys are not prone to hydrogen embrittlement except in the few instances where they contain cuprous oxide (namely tough pitch coppers and deoxidized coppers with low

residual deoxidizer). For aluminum and its alloys, it has been reported that hydrogen embrittlement is not a consideration in the presence of dry hydrogen gas but can be a problem under aqueous environmental conditions.

When considering polymers, research has shown that the most likely concern is over leakage at joints and valves in various points throughout the system, particularly in service lines and appliances. This is due to the increased permeability of hydrogen as compared to natural gas, caused mainly by the much smaller size of the hydrogen molecule. This increased permeability may also lead to a degradation in the stability of certain polymers in the presence of hydrogen.

Based on the findings in this literature review, standards for the various components used in gas delivery systems, such as check valves, shutoff valves, regulators, gauges, etc., as well as the relevant gas appliance standards, may need to be updated to account for the use of hydrogen or hydrogen/natural gas blends. This could include new or updated test methods to address material compatibility concerns with potential hydrogen blends.



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