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A review on hazards and risks to pipeline operation under transporting hydrogen energy and hydrogen-mixed natural gas

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Abstract. As an efficient and clean fuel, hydrogen energy plays an important role in relieving the energy crisis and achieving the orientation of zero carbon emissions. Transportation is the key link in the construction of hydrogen energy infrastructure. For large-scale and long-distance transportation of hydrogen, pipeline transportation has the advantages of high efficiency and cost saving. While using the existing natural gas pipeline to transport hydrogen, it would economize the economic cost, time cost and labor cost. However, the transportation of hydrogen may bring more hazards and risks. Based on the investigation of a large number of literatures, the research advance in hydrogen embrittlement, leakage, combustion and explosion risk of hydrogen and hydrogen-mixed natural gas pipelines was reviewed. The mechanism, research means and evaluation methods of hydrogen embrittlement, as well as the experimental and numerical simulation research results of leakage, combustion and explosion were discussed in detail. The definite and important conclusions include: (1) For buried hydrogen-mixed natural gas transportation pipeline, the leakage rate of hydrogen and methane is the same, the formation of the leakage crater is foreign to the nature of leakage gas. (2) When adding less than 25 volume percentage of hydrogen into the natural gas pipelines, the explosion risk would not be increased. Future research should focus on the risk prediction, quantitative risk assessment, intelligent monitoring, and explosion-suppression technical measures of hydrogen and hydrogen-mixed natural gas transportation pipelines, so as to establish comprehensive and multi-level pipeline safety protection barriers.

Keywords: Hydrogen embrittlement, Hydrogen leakage, Hydrogen-mixed natural gas pipeline, Risk assessment, Fire and explosion.

Abbreviations

AIDE	Adsorption-Induced Dislocation-Emission
BBN	Bayesian Belief Network
CCS	Carbon Capture and Storage
CERT	Constant Extension Rate Tensile
CFD	Computational Fluid Dynamics
CLT	Constant Load Test
DFT	Density Functional Theory
EBSD	Electron Back Scatter Diffraction
EHB	European Hydrogen Backbone
FEM	Finite Elements Method
FIB	Focused Ion Beam
HAC	Hydrogen-Assisted Cracking
HE	Hydrogen Embrittlement
HEDE	Hydrogen-Enhanced Decohesion
HELP	Hydrogen-Enhanced Localized Plasticity
HESIV	Hydrogen-Enhanced Strain-Induced Vacancies
HIP	Hydrogen Internal Pressure
HMT	Hydrogen Microprint Technique
IEA	International Energy Agency
LES	Large Eddy Simulation
LIST	Linearly Increasing Stress Test
MAA	Mesh Aluminum Alloy
NZE	Net Zero Emissions
R&D	Research and Development
SCC	Stress Corrosion Cracking
SEM	Scanning Electron Microscopy
SNM	Spherical Nonmetallic Material
SSRT	Slow Strain Rate Tensile
TEM	Transmission Electron Microscopy

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1 Introduction

Today, the world is in the first global energy crisis which was triggered by epidemic, war and other factors. In order to get out of the energy crisis, countries around the world are actively exploring new ways to replace fossil fuel energy, such as solar photovoltaic, wind energy, batteries, *etc.* This crisis would be a historical turning point of energy transition, from unrenewable energy to renewable, cleaner, and safer energy system. However, the swift development of new energy will also bring new challenges, especially before the maturity of recycling technology, the demand for critical minerals will increase so rapidly to cause fluctuations on mineral prices, as silicon and silver (used for solar photovoltaic), rare earth elements (used for wind turbine generators) and lithium (used for batteries) [1]. In addition to energy crisis, the human is facing another great challenge, which is global warming caused by greenhouse gas emissions, in which about 75% greenhouse gas from fossil fuels [2]. Therefore, for the sake of reducing the growth rate of global temperature, the amount of energy related carbon dioxide (CO_2) gas emissions should be cut down below a certain level. By 2050, if the CO_2 emissions fall to 32 Gallon ton (Gt), 12 Gt, and zero, the global temperature would rise to 2.5 °C, 1.7 °C, and less than 1.5 °C in 2100 respectively [1]. How does the world break away from today's crisis, and how to achieve Net Zero Emissions (NZE) by 2050? The answer is clean energy, with the biggest concern on the advanced batteries, hydrogen and the Carbon Capture and Storage (CCS). In addition to focusing on the Research and Development (R&D), an innovation of clean energy technologies, synchronous infrastructure is also crucial from now on to the next decade, especially for the construction of transport pipelines for hydrogen and captured CO_2 emissions [2].

Hydrogen is the lightest and most abundant element in the universe, ranking first in the periodic table of elements. However, there is very rare free state hydrogen on the earth and in the earth's atmosphere, mainly in the combined form, so hydrogen cannot be directly exploited, which is one of important secondary energy sources in 21st century. The global demand for hydrogen energy reached a record level in 2021, amounting to 94.3 million tons (Mt), which increased by 5.36% over 2020 (89.5Mt), by 3.4% over 2019 (91.2Mt) [3, 4]. Hydrogen has a wide range of uses, which can not only be directly used as combustion energy, but also be used as hydrogen-based fuels. At present, although the demand for hydrogen energy was concentrated in traditional application industries, the demand growth rate of emerging industries was growing rapidly. In the past three years, the hydrogen demand of the refining industry accounted for the highest proportion, reaching about 42%, while the hydrogen demand of the iron and steel manufacturing industry represented about 5%, and the demand of other emerging applications industries, such as transport, high-temperature heat in industry, power and buildings, only accounted for 0.4% of the total hydrogen demand, but increased by 60% in 2021 [5]. Taking the actual policies and measures into account, International Energy Agency (IEA) predicted that the global hydrogen demand would attain to 115Mt by 2030, and the new applications demand would reach nearly 2Mt [4]. The role of hydrogen in carbon neutralization has been widely recognized. Governments around the world have started to build new pipelines and reuse natural gas pipelines to transport hydrogen in their energy systems. For example, the European Hydrogen Backbone (EHB) estimated that the pipeline infrastructure for hydrogen transportation would be completed a pan-European network with a length of 53,000 km by 2040, in which the reused existing natural gas pipelines would account for more than 60% [6]. The earliest hydrogen pipelines in the world was in Germany (the Rhine-Ruhr metropolitan area), which had been in normal operation for 84 years. At present, there are about 5,000 km of hydrogen pipelines in the world, and more than 90% of hydrogen pipelines are laid in the United States and Europe [3]. The United States has the longest hydrogen pipeline, approximately 1600 miles (2576 km) [7], while Europe has about 2,000 km of hydrogen pipelines in operation [4]. In China, the first hydrogen long-distance pipeline was operated from Jiyuan to Luoyang. In 2021, the longest hydrogen transportation pipeline project in China was launched, with a total length of about 145 km, from Dingzhou to Gaobeidian, Hebei. The project is of great significance for solving the problem of hydrogen transportation from north to south in Beijing, Tianjin and Hebei region, forming regional hydrogen backbone network, and promoting the utilization of new energy in Beijing, Tianjin and Hebei region [8]. In the United States, hydrogen pipelines were operated by the company responsible for hydrogen transportation, but there was no common carrier company, which was different from natural gas pipelines [9]. Another big difference from natural gas pipeline was pipeline construction cost. From the comparison of construction cost breakdown for transporting methane, (CH_4), CO_2 and hydrogen (H_2) pipelines, the cost of CH_4 pipeline materials was the lowest, the cost of labor and right-of-way of CO_2 pipeline was the lowest, and miscellaneous cost of hydrogen was the lowest, while the total cost of CH_4 pipeline with the same diameter of 30 cm per kilometer was the lowest, and the total cost of hydrogen pipeline was the highest, about 1.2 times of the total cost of CH_4 pipeline [10]. The construction of pipelines to transport pure hydrogen was far less than the existing natural gas pipeline network. If the existing natural gas pipelines can be used to transport hydrogen, such as blending hydrogen into natural gas pipelines, the development of hydrogen energy will be of great benefit.

Consequently, from the beginning of this century, more and more countries in the world began to invest in the research of mixing hydrogen into the existing natural gas pipeline networks. In Europe, the Naturalhy project was started on 1st May 2004 and ended until 2009. The European Commission has adopted and funded the Naturalhy project, involving 39 European partners (15 from the gas industry) [11]. The Naturalhy project had completed the assessment of fire and explosion risk caused by hydrogen mixing with the natural gas and gas accumulation during the transportation of the H_2 and CH_4 mixture to the public, the impact of mixed gas on the reliability and integrity of pipeline materials and the effect of the presence of hydrogen on the use of mixture by end customers

[12]. In Germany, in 2013, the WindGas Falkenhagen project used wind energy electrolysis to produce hydrogen and methane, utilized natural gas pipe network for transmission of mixture of hydrogen and methane, explored the economic benefits of hydrogen energy brought by decarbonized assets [13]. In France, GRHYD project officially started at the 15th French Energy Conference in Dunkirk, which aimed to provide the power to housing units and buses in transportation by injecting hydrogen into natural gas pipelines and assess the technical and economic relevance [14]. In the UK, HyDeploy, the first practical hydrogen mixing project was launched in 2018, which aimed to provide the demonstration that the blending hydrogen was safe without changing the existing gas distribution network, the customers' behavior or the associated disruption. The project was divided into three phases, the first phase was to apply for permission to inject 20 mol% hydrogen into the natural gas pipeline, the second stage was the preparation stage before injection including the construction of electrolysers and other accessories and all training related to hydrogen injection, and the third stage was the live trial stage, where 20 mol% hydrogen blending gas was supplied for 100 households and 30 teaching buildings of Keele University [15]. In China, the first electrolytic hydrogen production and blending hydrogen with natural gas project was Chaoyang Hydrogen-mixed Natural gas Demonstration Project, which began in 2019, located in Chaoyang City, Liaoning Province. The project comprehensively verified the industrial chain of "production-storage and transportation- mixture-comprehensive utilization" for hydrogen, to break the foreign technology monopoly, to fill the gap in domestic specifications and standards for hydrogen blending in natural gas pipelines, and to promote the upgrading of relevant industrial systems [16]. On January 13, 2022, Enbridge Gas announced that the hydrogen mixing project was now fully operated in North America, which was described as the first hydrogen blending project [17]. The timeline of major hydrogen mixed natural gas pipeline transportation projects in the world is shown in Figure 1.

In the context of energy crisis and carbon neutralization, the development of renewable clean energy-hydrogen is very important. If hydrogen energy can be widely used, the precondition is to solve the problem of long-distance transportation. In addition to the direct construction of hydrogen transmission pipelines, the use of existing natural gas pipelines to mix hydrogen transmission is a key transition period for hydrogen energy application. However, as we all know, the physical and chemical characteristics of hydrogen determine the danger of hydrogen itself, especially the problem of metal hydrogen embrittlement, which would lead to the failure of metal pipelines and related equipment under hydrogen rich environment, leading to risks such as hydrogen leakage, combustion and explosion. It is very crucial to study the hazards and risks in the process of hydrogen transportation. It can also be seen from the research on the mixed hydrogen demonstration projects in various countries listed above. Therefore, the hazard of hydrogen induced failure of hydrogen pipelines, the risk of hydrogen leakage, combustion and explosion were mainly discussed from the aspects of hazard and risk generation mechanism, research methods and risk assessment in this paper, in order to comprehensively understand the potential difficulties of sustainable, safe and flexible energy pipeline industry in the new energy era.

2 Hydrogen embrittlement hazard

2.1 Mechanism of hydrogen embrittlement

The forms of Hydrogen Embrittlement (HE) to pipelines mainly included hydrogen induced cracking, hydrogen blistering, and hydrogen induced mechanical property changes [18–20]. In 1874, William H. Johnson, through three years of experimental observation, found that when the metal wire was immersed in dilute hydrochloric acid and dilute sulfuric acid for a few minutes, it would become more brittle. Through various experimental verifications, it was the occluded hydrogen that caused the change of metal properties after immersion in acid, and hydrogen would diffuse from half of the immersed acid to the other half that was not immersed in acid, leading to metal fracture [21, 22]. One hundred years later, in 1973, G.V. Karpenko, based on the existing theoretical and experimental data, proposed that the phenomenon of hydrogen embrittlement was regarded as a mechanical-chemical effects of metal selective microplasticizing caused by the chemisorption process of hydrogen activated by stresses [23]. However, until now, there has not been a unified mechanism that can explain the hydrogen embrittlement phenomenon.

2.1.1 Hydrogen-assisted cracking mechanism

Mechanism of Hydrogen-Assisted Cracking (HAC) is another common onym used in the all circumstances about hydrogen induced cracking, including delayed fracture, reductions in ductility, sub-critical cracking and fatigue cracking [24]. So far, the mechanisms to explain fracture of HE mainly include Hydrogen-Enhanced Decohesion (HEDE), Hydrogen-Enhanced Localized Plasticity (HELP), Adsorption-Induced Dislocation-Emission (AIDE), and Hydrogen-Enhanced Strain-Induced Vacancies (HESIV) [24–26]. HELP theory can be used to explain HAC in various metals and alloys [27]. In addition, other theories also proved that hydrogen can enhance dislocation mobility, such as using *in situ* Transmission Electron Microscopy (TEM) studies, macroscopic stress strain curves, and hydrogen shielding in elastic stress centers [28]. Hydrogen was considered to play a major role in promoting the engendering and accumulation of vacancies caused by deformation and in the HE sensitivity of high-strength steel [29]. Through further microscopic observation by Scanning Electron Microscopy (SEM), the flat feature on the surface of hydrogen induced fracture was revealed to have fine scale fluctuations related to the dense arrangement at the bottom layer of dislocations. Among the potential dislocation structures, hydrogen established dislocation substructure and local hydrogen concentration, which could increase the cracking tendency at specific locations [30]. Completely consistent with the HELP

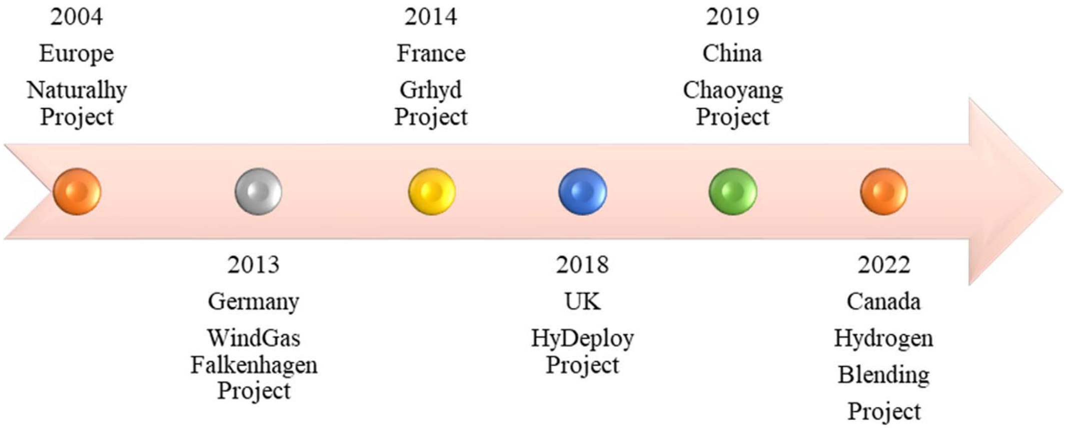

Figure 1. Timeline of major hydrogen mixed natural gas pipeline transportation projects.

process, the mechanism of hydrogen induced cracking was that the fracture surface resulted from the growth and merger of the voids that start at and extend along the slip band intersections, and the hydrogen assisted in the formation of the intense slip bands, reducing the stress required to produce initial voids, and accelerating the expansion of voids [31]. However, the mechanisms of HAC had been controversial. S. P. Lynch [32] questioned that the phenomena observed in literature [30] were explained only in the HELP mechanism framework, while ignoring the AIDE and HEDE mechanisms, and considered that its conclusion on fracture mechanism was unreasonable. The basis of AIDE and HEDE mechanism was that the hydrogen caused the weakening of metallic bonds, but no direct experimental evidence to support it [33]. In the past three decades, another hot spot in the study of HAC mechanism of steel and iron materials was the synergistic relationship between various mechanisms, especially the synergistic effect between HELP and HEDE, and HELP mediated HEDE [34]. Stan Lynch [35] proposed that the cracking mechanism AIDE, HELP and HEDE may occur simultaneously, and the fracture path and mode determined its main mechanism. AIDE was the main mechanism as cleavage fracture and dimple intergranular/transgranular fractures occur in steel; HEDE mechanism may be mainly used for brittle intergranular fracture; HELP mechanism was helpful for locating slip to cause slip zone fracture, but it may only play a secondary role in other fracture modes.

The mechanism of HESIV was hydrogen enhancement produced by strain induced vacancies. The damage evolution caused by vacancy aggregation was closely related to strong strain localization. The HESIV mechanism also plastically deformed at the stage prior to the initiation of cracking, and broke forward at the final stage to promote fracture [36].

2.1.2 Hydrogen blistering mechanism

Hydrogen blister was an important form of HE, which could induce the cracking of pipeline steel [19]. Hydrogen induced blistering could be considered as a special case of the pressure mechanism of HE [37]. The mechanism from hydrogen blistering to rupture can be considered as the Hydrogen Internal Pressure (HIP) mechanism caused by the increase of hydrogen pressure in the blister, and the hydrogen pressure may increase to several thousand atmospheres [38]. When hydrogen was filled in the metal material without cavities, holes containing hydrogen molecules of different sizes can be observed under TEM. When the HIP increased to equal to the critical stress of atomic bond fracture of the metal material, cracks would occur [39–41]. The effect of hydrogen blistering was increased with the increasing of hydrogen surface concentration and the trap density, and the trap density was increased as the increase of dislocation density [42].

2.2 Research method of HE

The research methods of HE can be divided into macroscopic experimental test, microscopic observation and molecular dynamics simulation, a new kind of numerical simulation. The test object was generally the metal charged with hydrogen, and the hydrogen charging modes included electrochemical hydrogen charging-to study the susceptibility to loss of ductility after cathodic hydrogen charging [43, 44], highly pressurized hydrogen charging-to study the effects of the hydrogen content under high-pressure hydrogen gas conditions on the HE of the steel [45–47], and gaseous hydrogen pre-charged [48]. The macroscopic performance of HE is the decline of toughness and plasticity of metal materials and the change of fracture form. Therefore, the impact of HE can be measured and evaluated by macroscopic mechanical experiments, including Constant Load Test (CLT), Linearly Increasing Stress Test (LIST), constant load Stress Corrosion Cracking (SCC) test, Constant Extension Rate Tensile (CERT) test and Slow Strain Rate Tensile (SSRT) test [48]. In terms of micro testing, the nano level mechanical testing of HE includes nano indentation experiment (the size of the indenter was about 100 nm to several microns), micro cylinder compression experiment and micro cantilever bending experiment, which the indentation was made into a micro cylinder or cantilever structure by Focused Ion Beam (FIB) technology, and based on the above experiments, the fracture mechanics parameters of the microstructure are determined, so, micro experimental research has a wide range of applications in the field of HE

Table 1. Qualitative results of hydrogen influence on four piping materials under different pressures and hydrogen concentrations [66, 68].

Pressure/bar g	Materials	H = <2 Vol.%	H = <5 Vol.%	H = <10 Vol.%	H = <100 Vol.%
<5.0	Steel	No effect expected	No effect expected	No effect expected	No effect expected H : Up to 20 Vol.%
<5.0	Stainless Steel	No effect expected	No effect expected	No effect expected	No effect expected
<5.0	Copper alloys	No effect expected	No effect expected	No effect expected	No short term effect expected
<5.0	Polyethylene	No effect expected	No effect expected	No effect expected	No effect expected
<8.0	Steel	No effect expected	No effect expected	No effect expected	No short term effect expected
<8.0	Stainless Steel	No effect expected	No effect expected	No effect expected	No short term effect expected
<8.0	Copper alloys	No effect expected	No short term effect expected	No short term effect expected	No short term effect expected
<8.0	Polyethylene	No effect expected	No effect expected	No effect expected	No effect expected
<10.0	Steel	No effect expected	No effect expected	No effect expected	Unknown
<10.0	Stainless Steel	No effect expected	No effect expected	No effect expected	Unknown
<10.0	Copper alloys	No effect expected	Unknown	Unknown	Unknown
<10.0	Polyethylene	No effect expected	Unknown	Unknown	Unknown
<60.0	Steel	Unknown	Unknown	Unknown	Unknown
<60.0	Stainless Steel	Unknown	Unknown	Unknown	Unknown
<60.0	Copper alloys	Unknown	Unknown	Unknown	Unknown
<60.0	Polyethylene	Unknown	Unknown	Unknown	Unknown

research [44, 49–59]. The diffusion process of hydrogen in pipelines is a prerequisite for the development of HE failure [60]. The hydrogen permeation distribution can be studied by Hydrogen Microprint Technique (HMT) and Electron Back Scatter Diffraction (EBSD), the former can realize online observation and visualization [61, 62], while the latter can study the influence of microstructure on hydrogen diffusion [60]. However, the above research techniques can only show the planar distribution of hydrogen penetration, and cannot directly observe the three-dimensional distribution of hydrogen in the metal. At present, generally, numerical simulation techniques are used to establish the three-dimensional distribution model of hydrogen in the metal, mainly including Finite Elements Method (FEM) and Density Functional Theory (DFT) [63]. In addition, the combination of DFT and molecular dynamics simulation technology can also establish the interaction model between hydrogen and metal [64, 65].

2.3 Assessment the effect of HE on pipeline performance

There are many parameters that can be used to evaluate the sensitivity of materials to HE. The test measures used to evaluate the degree of HE include: reducing the failure strain and fracture stress, extending the failure time, and reducing the fracture energy of hydrogen containing materials [35]. However, if the impact of HE on material strength, ductility and fracture toughness was evaluated only by laboratory tests, then the results of hydrogen impact usually were very different, due to the factors including different sample designs, experimental designs, bad practices and data dispersion [66]. Andrews *et al.* [67] applied the industry standard model for the plane dents, volumetric corrosion, parent metal cracks and fatigue crack growth damage of European onshore transportation pipelines, and predicted the modern high toughness pipes and the old low toughness pipes. The results showed that although the high toughness materials had degraded under the hydrogen exposure environment, they could withstand the damage, while the low toughness materials could not withstand serious damage. In addition to evaluating the performance impact of HE on pipeline materials, it is also very important to evaluate the maximum allowable hydrogen content of existing natural gas transmission pipelines, but it is difficult to draw a clear conclusion for all pipelines. The draft CEN/ISO report [68] listed the results of the effects of hydrogen on four different pipe materials when mixing different proportions of hydrogen concentrations at different pressures, and the results were no effect expected, no short term effect expected and unknown respectively as shown in Table 1.

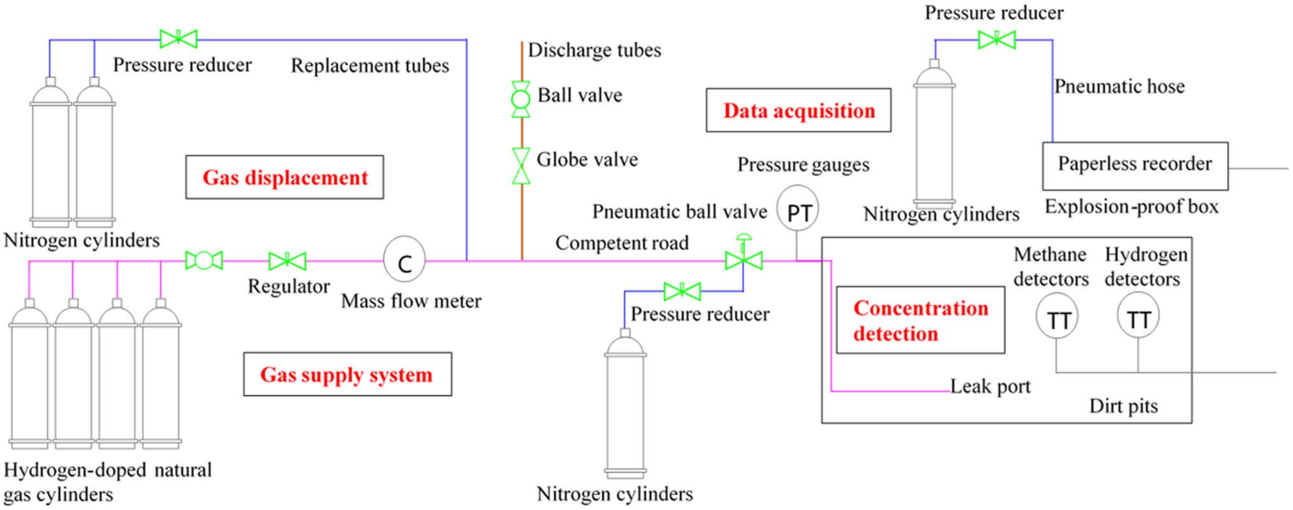

Figure 2. Underground pipeline leakage test system for hydrogen-doped natural gas [76].

As described in the above information, the maximum allowable amount of hydrogen mixed in the existing natural gas pipeline depends on the pipeline transmission pressure, pipeline material and other operating parameters. In some existing standards and guidelines, the maximum allowable hydrogen concentration mentioned includes 10%, 15%, 20% and 100% [68–73].

3 Hydrogen leakage risk from pipeline

3.1 The research of hydrogen leakage

Because hydrogen molecules are much smaller than methane, the public is much more worried about the risk of hydrogen leakage from both hydrogen pipelines and hydrogen mixed natural gas pipelines. Except for unexpected situations, such as irresistible disasters, operational errors, and man-made vandalism, most of the leakage occurs in loose threaded joints, damaged sealing materials, and cracked pipe walls [74]. Compared with the joint connection and seal, the pipe has a larger surface area, therefore, the leakage through the pipe wall is the main cause of gas loss in the system [70]. Research methods of hydrogen leakage mainly include experimental method and numerical simulation method.

The experimental measurement is usually to measure the hydrogen leakage flow rate and hydrogen concentration distribution under different pressures and temperatures through pinholes of different diameters, in which the hydrogen leakage flow rate is the most important factor to evaluate the impact of hydrogen leakage [74]. The diffusion mechanism of hydrogen leaking jet from pinhole nozzle is studied by using shadow flow visualization in the experimental scene [75]. The more similar the experimental scenario is to the actual situation, the more consistent the experimental results would be with the actual situation. Therefore, Zhu *et al.* [76] developed a large-scale experimental system that can simulate the hydrogen mixed natural gas pipeline buried in soil, and carried out the experimental measurement of the leakage and diffusion of hydrogen mixed natural gas under different hydrogen mixing ratio, release pressure and leakage direction. The schematic diagram of the full-scale experimental test system is shown in Figure 2.

In the leakage experiment from hydrogen mixed natural gas pipeline, it is concluded that hydrogen will not leak preferentially and the leakage rate of hydrogen is equal to that of natural gas [77]. For the leak location given in the experiment, if the flow rate was higher, the dispersion rate of hydrogen in space would be faster [78, 79]. The differences between different leak locations depend on the flow, and the flow rate determines the degree of hydrogen dispersion rather than the leak location [79]. To the buried pipelines, Deborah *et al.* [80] verified through full-scale experiment that the crater formation was related to the gas leakage pressure, the release direction, and the nature of the soil hosting the pipeline, but not to the nature of the leaked gas. The schematic diagram of gas released from crater formation is shown in Figure 3.

The models used in the numerical simulation study of pipeline hydrogen leakage include Computational Fluid Dynamics (CFD) models and the Large Eddy Simulation (LES) model. The software used in the numerical simulation study of hydrogen leakage is mainly ANSYS Fluent software. At present, there are parallel CFD programs, such as GASFLOW-MPI developed by Karlsruhe Institute of Technology in Germany, which has been used to predict the flow [81], diffusion [82, 83], combustion and explosion of hydrogen and other gases [84]. Li *et al.* [85] carried out numerical simulation on the leakage of hydrogen-mixed natural gas, and found that the higher the proportion of hydrogen-mixed natural gas, the longer the duration of the dangerous area after the leakage. Olvera and Choudhuri [86] used standard k-e turbulence model to compute dispersion patterns of hydrogen and methane and found that the potential risk of hydrogen was greater, but with the rapid rise of hydrogen, the impact on the release of surrounding buildings was rapidly reduced. Tang *et al.* [87] studied the

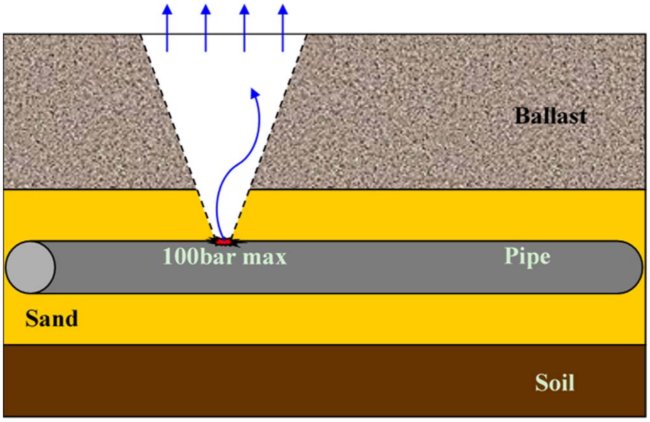

Figure 3. Crater formation giving free gaseous release on a buried pipeline [80].

instantaneous and average hydrogen concentration distribution of the under-expansion hydrogen jet stream under a storage pressure of 82 MPa from a small orifice with a diameter of 0.2 mm using three-dimensional (3D) numerical calculations. The numerical simulation technology can dynamically analyze and calculate the changes of density, diffusivity, viscosity and flammability zones in the process of hydrogen leakage and diffusion from the pipelines [88, 89]. The scene of transporting mixture gas into the actual kitchen through hydrogen-blended natural gas pipeline can still be simulated by ANSYS fluent software to analyze the influence of hydrogen blended ratio, leakage rate, ventilation conditions and size on the leakage and diffusion characteristics and evolution [90]. In addition, the numerical simulation technology can simulate the leakage of hydrogen or the mixture of hydrogen and natural gas in various scenarios, such as compressor plant, mobile hydrogen refueling station, *etc.*, and can analyze the impact of leakage flow, leakage location, leakage orifice diameter, pressure and wind speed on the diffusion range [91, 92].

In addition, the measured experimental data can be used to calibrate commercial CFD models and verify the accuracy of numerical simulation results, which are often used to study hydrogen release and diffusion [93]. Wang *et al.* [94] used schlieren and high-speed camera to obtain the jet shape of hydrogen leakage of the double ferrule joints, which was consistent with the results of the calibrated CFD model simulation. Malakhov *et al.* [95] used a part of the underground mine tunnel to carry out hydrogen leakage experiment, and obtained the hydrogen distribution with different inlet pressures and different leak orifice sizes through the sensors around the leak orifice. However, in some complex cases, there were quite large deviations between the experimental results and the numerical simulation results. Due to the explosive characteristics of hydrogen, helium is often used to replace hydrogen in the laboratory to ensure the safety during the experimental process. Shu *et al.* [96] conducted helium (instead of hydrogen) leakage dispersion experiment in the environmental chamber to verify the accuracy of the simplified prediction model. He *et al.* [93] used the small-scale experiment of measuring helium concentration to verify the current CFD model, and then ran the same CFD model using the physical property value of hydrogen, thus transforming the helium experiment into hydrogen diffusion simulation under different scenarios.

3.2 Hydrogen leakage risk assessment

For specific transportation pipelines, many different leakage events may occur due to the different leakage location, leakage flow rate, leakage flow rate and leakage direction, and the severity of the leakage and the final damage consequences also have many possible ways. Therefore, the assessment and prediction of leakage and consequences through associated variables are random in nature [97]. Pasman and Rogers [97] used the Bayesian Belief Network (BBN) software to model the hydrogen distribution and transportation mode, and concluded that the leakage risk of liquefied hydrogen was greater than that of compressed gaseous hydrogen. Kodoth *et al.* [98] estimated hydrogen leakage rate by time series method, leakage hole size and nonparametric method (based on Bayesian update), and the estimated data of the three methods had similar trends, in which the nonparametric method was the most conservative.

3.3 Detection methods of hydrogen leakage

Due to the high combustibility and large explosion range of hydrogen, once leakage occurs, it is easy to cause large combustion and explosion accidents, so it is very necessary to detect hydrogen leakage to ensure safety. Zou Qiang *et al.* [99] proposed a hydrogen leakage detection method based on support vector machine, and the correct rate of identifying small flow hydrogen leakage was higher than 90%. Miao Yang *et al.* [100] proposed a fast visual detection method for high-pressure hydrogen leakage, using digital image processing technology to detect hydrogen leakage. Falsafi *et al.* [101] developed a hydrogen leakage sensor based on Sm-doped cobalt ferrite ($\mathrm{Smx{-}CoFe_2{-}xO_4}$) material, and tested that the sensor had good sensitivity, stable behavior, fast response and recovery ability. When hydrogen is mixed with natural gas, if the gas detector cannot maintain sufficient sensitivity to the mixed gas, the gas detector may cause incorrect readings. Sun *et al.* [102] developed a fast response colorimetric hydrogen detector based on $\mathrm{Pd/MoO_3}$ nanocomposites, which can show rapid and strong color change at the exact hydrogen leakage location, and can easily monitor the hydrogen leakage. Hall *et al.* [103] analyzed the reaction of various natural gas industrial detectors to hydrogen mixture up to 20 vol% in natural gas as part of the HyDeploy project and found that a few measurement ranges of multiple detectors commonly used in the natural gas industry have different degrees of cross-sensitivity to hydrogen in the natural gas mixture. Blokland *et al.* [104] developed a chip coated with platinum nanocomposite layer, which was added to the natural gas sensor in order to expand the applicability to the detection of mixing with hydrogen, and found that the sensor with this chip can accurately and reversibly detect the hydrogen concentration in the mixed gas, so as to facilitate quality and process control.

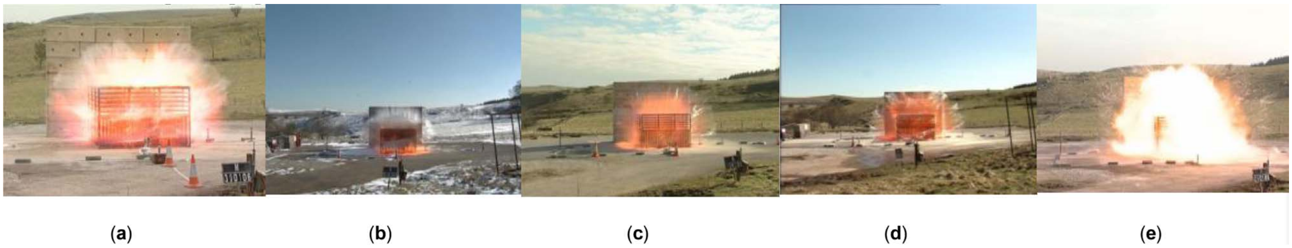

Figure 4. Explosion image after ignition with different hydrogen doping ratio [115]. The ratio is list as: (a) 0% hydrogen; (b) 25% hydrogen; (c) 51% hydrogen; (d) 75% hydrogen; (e) 100% hydrogen.

4 Fire and explosion of hydrogen and hydrogen-blended natural gas pipelines

4.1 Hydrogen fire research

The main risks of pipelines transporting hydrogen are the thermal radiation of continuous fire with the impact pressure of gas cloud explosion, and the fire hazard area is slightly greater than other events [105]. In the experimental study of hydrogen flame, Schefer *et al.* [106] recorded the flame with video to estimate the flame length and structure, and used a radiometer to measure the characteristics of radiant heat flux. Mogi and Horiguchi [107] found from experiments that the flame size was related to the nozzle diameter and the release pressure and the radiation was predicted according to the gas flow rate and the distance from the flame. Mogi *et al.* [108] confirmed that the spontaneous combustion frequency of the hydrogen jet raised with the increasing of the discharge pipe length in experimental research. Friedrich *et al.* [109] studied the flame stability and propagation behavior through hydrogen jet combustion experiments. From two large-scale experiments of the Naturalhy project, Lowesmith and Hankinson [110] found that when operating under the same pressure, the pipeline transporting hydrogen-blended natural gas caused slightly less harm in terms of heat dose than the natural gas pipeline. Panda and Hecht [111] measured the maximum ignition distance by using non-invasive laser spark focused on the jet axis, and obtained that the ignition distance was linearly proportional to the effective injection diameter. Mei *et al.* [112] carried out numerical research on flame propagation during hydrogen explosion in bend pipes with different angles, showed that the influence of bend structure on flame propagation was mainly concentrated in the middle and late stages.

4.2 Hydrogen explosion research

Takeno *et al.* [113] clarified through experiments that the explosion power depends on the concentration and volume of hydrogen/air premix and the turbulence characteristics before ignition. Lowesmith *et al.* [114] conducted a series of large-scale explosion experiments involving methane/hydrogen mixture in the Naturalhy project, which showed that when adding 20 vol% hydrogen to methane would lead to a small increase in the explosion flame speed and overpressure, and when adding 50 vol% hydrogen, it would increase significantly. Under the condition that the congested area of 3 m × 3 m × 2 m contains multiple layers of pipes, Royle *et al.* [115] conducted ignition experiments on the composition of the methane/hydrogen mixture used between 0% hydrogen and 100% hydrogen, and the results showed that the explosion overpressure generated by the methane/hydrogen mixture with 25 vol% was much less than those generated by methane alone. After an interval of 11 years, Shirvill *et al.* [116] repeated the above experiments and obtained similar research results, and concluded that adding 25 vol% of hydrogen to the pipeline network would not significantly increase the risk of explosion. The explosion images after ignition under various conditions with the ratio of hydrogen mixed with natural gas ranging from 0 to 100% are shown in Figure 4.

Wang *et al.* [117] conducted experiments and simulation research to show that the jet fire and explosion caused by hydrogen spontaneous combustion must go through a series of specific development stages, and this process has no significant correlation with pipeline length and release pressure. Zhou *et al.* [118] established a CFD three-dimensional simulation model to study the consequences of hydrogen-doped natural gas leakage and explosion in urban pipeline, and the results showed that the traffic flow changed the diffusion path of the jet, the flammable gas cloud formed a complex contour in many obstacles, and the high congestion level led to more serious explosion accidents.

4.3 Research on prediction and assessment of hydrogen fire and explosion

In most cases, the engineering model originally developed for natural gas explosion can be modified and applied to predict the hazards caused by explosion involving natural gas/hydrogen mixture [119]. Zhao *et al.* [120] designed a fire risk grade prediction program for hydrogen pipeline and carried out fire risk grade prediction analysis, and the results showed that the finite Ridgelet neural network optimized by the improved firefly algorithm has advantages in prediction accuracy and effective of the safety status of hydrogen pipeline. Froeling *et al.* [121] analyzed the personal risks related to the dangerous hydrogen jet fire using the flame model and calculation software, and concluded that the level of lethality generated by the hydrogen jet fire was lower than that of the natural gas jet fire. Russo *et al.* [122] evaluated the damage of high pressure hydrogen pipeline explosion to building structure with pressure pulse diagram signify XXXX. Based on the utility theory and the ELECTRE TRI method, Viana *et al.* [123] proposed a dimensional risk model and a probability model, and classified the hydrogen pipeline sections according to the risk level, so as to bring the risk analysis elements into the assessment of unexpected situations, probabilities and consequences. Li *et al.* [124] used the CFD method to establish a three-dimensional model to simulate the diffusion behavior of hydrogen-containing natural gas released from the transmission pipeline, predict the development process of combustible gas cloud, and evaluate the dangerous area generated by the hydrogen-natural gas mixture.

4.4 Research on fire and explosion suppression measures

Engineering explosion suppression materials and structures can be used to mitigate hydrogen explosion in pipelines. The explosion suppression material is a network or porous structure filled into the pipeline, which reduced the flame speed and hydrogen concentration through specially designed shape and size, thus alleviating the explosion effect [125]. However, the materials and structures that can inhibit the methane gas explosion may not have a satisfactory effect on the hydrogen explosion. Pang *et al.* [126] experimental research showed that the Mesh Aluminum Alloy (MAA) product not only could not effectively inhibit hydrogen deflagration, but also increased the maximum explosion pressure, which was contrary to the satisfactory inhibition effect of MAA product on methane deflagration. Song *et al.* [127] found that Mesh Aluminum Alloys (MAAs) and Spherical Nonmetallic Materials (SNMs) have dual effects of promoting/inhibiting hydrogen deflagration, and SNMs show better inhibition effect than MAAs. Porous coatings and porous materials (such as polyurethane foam, steel wool) that can attenuate shock and detonation waves can also be used as suppression materials for hydrogen explosion or hydrogen and methane mixture explosion [128–130]. Besides materials and structures, there are other methods to suppress fire and explosion. Wen *et al.* [131] found that the ultra-fine water mist has a significant mitigation effect on the explosion of hydrogen/methane mixture in the ventilated room with obstacles, and the mitigation effect will also improve with the enhancement of water mist flow. The existence and location distribution of obstacles are the key factors to determine whether the explosion suppression effect is achieved. Xia *et al.* found [132] that when there was no obstacle, 8 mm, 15 mm and 30 mm water mist can significantly reduce the flame velocity and explosion overpressure of hydrogen, and 45 mm water mist had no inhibition effect, however, when the water mist was released near the obstacle, the results were completely opposite. Duan *et al.* [133] found that when obstacles were symmetrically distributed, porous materials with different thickness would promote or suppress explosion flame and overpressure.

5 Conclusion

Since 2020, due to epidemic, war and other factors, the world is facing the first energy crisis. On the other hand, with the increasing global warming, in order to achieve the goal of zero carbon emissions by 2050, the global energy departments are shifting from fossil fuel energy to renewable clean energy, and hydrogen energy plays a crucial role. At present, hydrogen energy has been used for many purposes, but large-scale use of hydrogen energy must solve the problem of efficient transportation from the production to the client, and pipeline transportation is the first choice. However, due to the nature of hydrogen, the pipeline transportation of hydrogen and hydrogen/methane mixture will pose new threats and risks. Based on a large number of literature research, this paper summarizes the research advance of hydrogen embrittlement, gas leakage, combustion and explosion hazards and risks of hydrogen and hydrogen mixed natural gas pipelines. The summary and prospect are as follows:

1. Hydrogen embrittlement is the main hazard of hydrogen and hydrogen-mixed natural gas pipelines. The transportation of hydrogen pipeline needs to be carried out under pressure. Under high pressure, hydrogen molecules will produce adsorbed hydrogen atoms on the surface of pipeline steel and enter the steel, causing hydrogen embrittlement and endangering the safe operation of the pipeline. Comparing and summarizing the research results from three aspects: the mechanism, the research methods and the evaluation. The experimental verification supporting the theoretical mechanism is still lacking. Although the evaluation of material hydrogen embrittlement by macro and micro experiments and numerical simulation is relatively mature, it is difficult to accurately serve in the actual project. Accurate prediction and quantitative evaluation of the structural integrity and service life of the pipe wall, joint and important components of the actual transportation pipeline will become important research direction for the safety of hydrogen transportation in the future.
2. Hydrogen leakage may be caused by hydrogen embrittlement or accidental conditions, which may lead to major accident risks such as hydrogen combustion and explosion. In response to the public's concern that hydrogen is more likely to leak than methane, a full-scale simulation experiment was conducted on the buried hydrogen-mixed natural gas pipeline, and it was concluded that hydrogen will not leak preferentially, and the leakage rate is the same as methane. The formation of the leakage crater is related to the pressure, direction and soil properties, and has nothing to do with the nature of the leakage gas. The risk assessment of hydrogen leakage is mainly based on Bayesian method. The detection technology of hydrogen leakage includes vector machine detection, digital image processing technology, composite materials and coating chips.
3. Research on hydrogen fire focused on flame study exploring factors such as flame length, structure, radiant heat flux, spontaneous combustion frequency, ignition distance, and the impact of bend structure on flame propagation. Meanwhile, the experimental study of hydrogen combustion and explosion showed

that when the proportion of hydrogen injected into the natural gas pipeline is less than 25%, the explosion risk will not be significantly increased. The research of explosion suppression measures mainly focuses on materials, structures, obstacles and water mist. Future research should focus on revealing the evolution laws of risks such as leakage, combustion and explosion under different hydrogen mixing ratios, developing comprehensive and accurate quantitative risk assessment technology, intelligent monitoring leakage technology and explosion-suppression technology for hydrogen transmission pipelines.

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Conflicts of Interest

The authors declare no conflict of interest.

Author contributions

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