



Caveats of green hydrogen for decarbonisation of heating in buildings

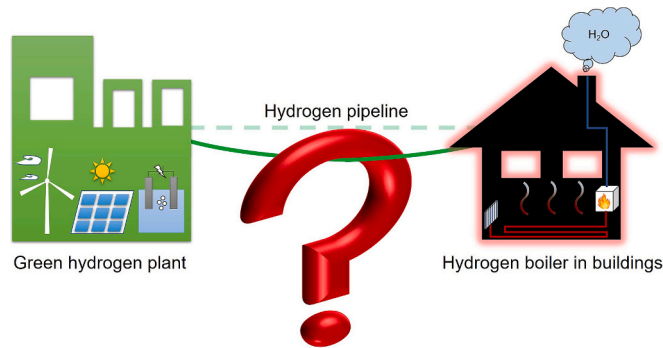
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HIGHLIGHTS

- Whole energy landscape for heating in buildings by green hydrogen is discussed.
- Relevant and recent government reports and research articles are reviewed.
- Appropriate water resourcing and plant sizing are essential to realising the goal.
- HENG containing 20–23 vol%H₂ is attainable using the existing infrastructure.
- 100% H₂ can eliminate the emissions, but CO, C_xH_y, and UHC may be emitted by HENG.

GRAPHICAL ABSTRACT



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ABSTRACT

Hydrogen (H₂) has rapidly become a topic of great attention when discussing routes to net-zero carbon emissions. About 14% of CO₂ emissions globally are directly associated with domestic heating in buildings. Replacing natural gas (NG) with H₂ for heating has been highlighted as a rapid alternative for mitigating these emissions. To realise this, not only the production challenges but also potential obstacles in the transmission/distribution and combustion of H₂ must be technically identified and discussed. This review, in addition to delineating the challenges of H₂ in NG grid pipelines and H₂ combustion, also collates the results of the state-of-the-art technologies in H₂-based heating systems. We conclude that the sustainability of water and renewable electricity resources strongly depends on sizing, siting, service life of electrolysis plants, and post-electrolysis water disposal plans. 100% H₂ in pipelines requires major infrastructure upgrades including production, transmission, pressure-reduction stations, distribution, and boiler rooms. H₂ leakage instigates more environmental risks than economic ones. With optimised boilers, burning H₂ could reduce GHG emissions and obtain an appropriate heating efficiency; more data from boiler manufacturers must be provided. Overall, green H₂ is not the only solution to decarbonise heating in buildings, and it should be pursued abreast of other heating technologies.

1. Introduction

This paper explores the challenges and opportunities associated with

the adoption of hydrogen for heating from an end-to-end supply chain perspective, *i.e.*, from production to distribution to end-use of hydrogen for heating.

About 14% of CO₂ emissions globally are caused by using fossil fuels

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Nomenclature*Abbreviations and chemical formulas*

| | |
|------------------|--|
| AEM | Anion exchange membrane |
| API | American petroleum institute |
| BWRS | Benedict–Webb–Rubin–Starling equation of state |
| CapEx | Capital cost including the initial investment |
| C_xH_y | Hydrocarbon side products |
| CO | Carbon monoxide |
| CO ₂ | Carbon dioxide |
| GHG | Greenhouse gas |
| GWP | Global warming potential |
| H ₂ | Green hydrogen |
| HENG | Hydrogen-enriched natural gas |
| HHV | Higher heating value |
| IGT | Institute of Gas Technology in the United States |
| LBV | Laminar burning velocity |
| LHV | Lower heating value |
| NG | Natural gas |
| N ₂ O | Nitrous oxide |

| | |
|-----------------|--|
| NO _x | NO and NO ₂ greenhouse gases |
| NTS | National transmission system for natural gas in the United Kingdom |
| OpEx | Operational costs including maintenance, etc |
| PCCE | Proton conducting ceramic electrolyser |
| PEM | Polymer electrolyte membrane, Proton exchange membrane |
| RE | Renewable electricity |
| SOEC | Solid oxide electrolyser cell |
| STP | Standard temperature (0 °C) and pressure (1 atm) |
| UHC | Unburnt hydrocarbons |

Greek letters

| | |
|------------|---------------------------|
| Z | Compressibility factor |
| I_W | Wobbe index |
| λ | Air-to-fuel ratio |
| μ_{JT} | Joule-Thomson coefficient |
| ρ | Density |
| ϕ | Equivalence ratio |

such as natural gas (NG) to generate heat in buildings for domestic hot water and space heating [1,2]. Currently, there are two major approaches to abate these emissions: hydrogen boilers and heat pumps. The advantage of the former is that it does not fully rely on the electricity supply and it allows for offsetting the intermittency of the electrical power, whether it is from renewable sources or conventional polluting power plants. Although the latter offers a high energy conversion rate (or coefficient of performance) of up to 400%, the principal challenge with heat pumps is their upfront cost which can be quite costly and even prohibitive [3]. As many households are already connected to the NG grid and have a gas boiler installed (e.g., 90% in England [4]), it is deemed logical to propose the use of the same infrastructure for H₂ to eliminate heating-related carbon emissions. In recent months, the controversy around the use of H₂ for heating in buildings has reached the news headlines that can drastically shape or reshape public opinion when discussing a new technology; a consumer can be easily persuaded when reading “Hundreds sign up for Fife’s world-first hydrogen homes project to heat houses!” [5] or dissuaded when reading “Hydrogen boilers might need ‘four-inch holes in walls to prevent explosions!’” [6]. To counteract this unnecessary, and perhaps contentious issue in the heating industry, we have been prompted to look at the available data on different aspects of the H₂ supply chain for heating in buildings.

To the best of our knowledge, no study deals with the holistic technical practicality of green H₂ as a short/long-term route for the decarbonisation of heat. A multidisciplinary discussion around Hydrogen is carried out in [7] with a focus on hydrogen production and storage technologies and transport end-use. The study highlights the need for further multidisciplinary research on domestic hydrogen. The impact of hydrogen on heating is another aspect considered in the literature [8]. However, they tend to focus on hydrogen production technology and increased electricity demand arising. A recent study developed a socio-technical framework to identify the barriers to the adoption of hydrogen in domestic settings [9]. However, the focus is on interactions between social and technical barriers with discussions on hydrogen pathways and excludes the technical barriers associated with end-use, *i.e.*, combustion. Thus, this review aims at collating the available state-of-the-art data and providing more up-to-date information on the known caveats when discussing H₂ or H₂ blends in pipelines and boilers for heating in buildings (residential and/or commercial). To this end, recent reports and case studies were investigated in detail to shed light on the feasibility of commercially discussed routes to an H₂-driven heating landscape. The topics discussed here encompass three principal areas:

resource requirement for green H₂ production, H₂ or H₂-enriched NG (HENG) blend transmission/distribution *via* NG infrastructure and combustion in - boilers. For scaling purposes, some examples from the United Kingdom and the United States are derived to paint a clearer picture of the current status and the requirements associated with meeting net-zero goals.

As the focus is on decarbonisation, this paper only considers green H₂ production. The state of the art of different hydrogen production technologies is already widely discussed [10] and further developments in green hydrogen production such as salt water splitting [11] are progressing at a rapid pace. The water requirements for electrolysis, cooling and blow-out losses are estimated for the production of hydrogen sufficient to meet the heating demands of the UK. This estimate is still significantly less than the water requirements for fossil fuel plants. However, further policies and regulations are required with regard to highly mineralised wastewater disposal.

Repurposing existing infrastructure can - theoretically - offer a rapid and low-cost route to the decarbonisation of heat. However, there are challenges such as material stability and equipment compatibility with such a small and light molecule, as well as with the combustion of this highly volatile gas. Furthermore, as H₂ is a quantum gas (thermodynamically behaves differently than most gases [12]), the equipment for the measurement of the flow rate, temperature and pressure could require retrofitting or replacement. This could create new logistic and economic challenges when maintaining and inspecting the H₂ lines instead of NG pipelines.

On the other hand, we also face the challenge of optimised combustion to minimise the formation of C_xH_y, CO and/or NO_x when burning HENG or pure H₂ in a boiler. Understanding H₂ combustion is of great significance, as the main objective of using H₂ instead of NG is emission abatement. Hence, side production of other greenhouse gases (GHGs) would (at least partially) defeat the purpose.

2. Production

In this section, we focus on the requirements for water, renewable electricity (RE) and other resource management. We use the UK as a case study to discuss the resource challenges to adopting hydrogen for heating in homes.

2.1. Water

Domestic consumption accounts for the largest use case of NG in the UK [13]. In 2021, this amount was recorded at 318.4 TWh of domestic NG use. Assuming 48 kWh of RE and 21 L of fresh water are needed to produce 1 kg of H₂ [14], this is equivalent to 9.6 Mt. of H₂, requiring about 459 TWh of RE and 201 Mm³ of water for production *via* electrolysis, excluding the power required for water treatment. For scale, the amount of water required for hydrogenising domestic heating in the United Kingdom alone amounts to 2.6 times the volume of water in the Caspian Sea – the largest inland body of water in the world. If water is extracted at this amount annually, there are concerns about environmental consequences when seawater is desalinated at such a scale, and all the vapour from burning H₂ is put back into the natural water cycle. However, C. Sherwood et al. [15] investigated the effect of the latter on global warming and concluded that this effect is very small or in some cases negative due to the cooling effect of near-surface water vapour.

In addition to the water for electrolysis, larger electrolyser stacks including megawatt- and gigawatt-scale plants will introduce new water requirement loads in the form of the evaporative cooling side and blow-out losses. Although rather pessimistic, 60–95 L/kgH₂ of water has been reported when accounting for the use of saltwater coupled with reverse osmosis desalination as feedstock for green H₂ [16]. Overall, R. R. Beswick et al. [17] estimate that green H₂ production consumes 1.5 ppm of global freshwater or 30 ppb of saltwater. They state that this estimate is 33% lower than the water consumed by fossil fuel energy production and power generation, and therefore, the numbers presumed in their study show a clear case for replacing fossil fuels with green H₂. However, based on our calculations, they have not normalised these values by the amount of energy produced. Based on publicly available data and the amount of water mentioned in this paper for the future production of H₂, the water consumption per unit of energy produced (HHV-based) is roughly similar between the fossil fuels (0.24 L_{water}/kWh_{HHV}) and H₂ (0.23 L_{water}/kWh_{HHV}). It should be also noted that the water withdrawal for H₂ production would be ≥ 0.53 L_{water}/kWh_{HHV}, which is much lower than 1.95 L_{water}/kWh_{HHV} of fossil fuels. The following assumptions were made for the above calculations:

- Amount of water for future hydrogen: 20.5 bcm [17]
- 2014 fossil fuel water consumption: 31 bcm [17,18]
- 2014 fossil energy production: 128,770 TWh [19] (fossil fuels: coal, oil, and natural gas [20])
- Water withdrawal for electrolysis: 21 L/kgH₂ [14]

Furthermore, through our validation exercise for the findings of [17], it was found that fossil fuels refer to the production of coal, oil, and natural gas [17,19,20], and power production refers to the electricity generation in conventional thermal power plants burning coal, oil, and natural gas [17]. Therefore, this water consumption also accounts for the water required for generating useful work as electricity, which is mainly driven by water for cooling requirements. To account for the water consumed to produce useful work as electricity *via* a fuel cell stack, we have made the following assumption that fuel cell cooling is identical to electrolyser cooling with no heat recovery system and CHP integration. Accordingly, a fraction of water requirement to produce electricity from H₂ using a fuel cell stack with an overall energy efficiency of 50% (rated efficiency [21]), would be negligible and the overall water withdrawal value of 1.50 L_{water}/kWh_{HHV} remains and may go up to 1.70 L_{water}/kWh_{HHV} depending on the cooling strategy [16]. It is anticipated that this withdrawal will be even lower than 1.50 L_{water}/kWh_{HHV} in a hydrogen future where cooling systems are also more advanced and efficient. To conclude, the water withdrawal in the future hydrogen scenario would still remain less than the current withdrawal for the energy sector.

From an individual production site perspective, not only sizing but siting of electrolysis plants must also be considered as a vital part of the

Water-Energy Nexus [22]. Appropriate resourcing of water will influence the levelised cost of green H₂ produced and of course, determine whether the feedstock is sustainable. For example, if wastewater from industry is used as feedstock, this could dramatically reduce the cost of feedstock and transmission if the plant is located nearby [16] [23]. For instance, potential H₂ generation from an average industrial wastewater site is reportedly in the order of 1000 TWh/yr [24].

Even with proper sizing and siting of such plants, however, there still remains the question of what to do with the highly mineralised water after electrolysis. Some of the modern techniques for purification of wastewater are electrocoagulation [25], nanofiltration [26], biosorption [27], and advanced oxidation [28]. Nonetheless, this introduces another energy-consuming aspect: to sustainably dispose of the post-purification hard water in the electrolysis plants (e.g., 0.511 kWh/m³ [29]). Unfortunately, safe sludge disposal has eluded the recent studies on the use of wastewater for green H₂ production [23,30]. Currently, aerobic and/or anaerobic digestion is a common method for converting organic solids to more inert compounds. However, using industrial wastewater as feedstock for green H₂ is expected to result in higher concentrations of non-organic compounds and heavy metals in the sewage. For the time being, it is believed that most reports assume the traditional recycling and reusing protocols such as in [31] can be applied to electrolysis-related wastewater. Considering the expected scale of green H₂ production, however, such policies and protocols may require revision and modification during and after the transition to an H₂ economy.

2.2. Renewable electricity

It is important to reflect on the application of H₂ in the first place, which is to store RE and mitigate its intermittency. Therefore, green H₂ is likely to be produced in a high-wind season for example, which inhibits the spread of production throughout the year to minimise the environmental impact of this endeavour. Another challenge of producing green H₂ in such high capacity is the available RE farms which are mainly offshore. Estimates are about 8 GW of global electrolysis capacity in 2022 [32], which translates into an annual 64 TWh of RE assuming 8000 h of yearly operation. One can see the dramatic discrepancy between the current state of electrolysis plants and what is required to provide the domestic sector (just UK: ≥ 459 TWh).

2.3. Other resources

The effect of electrolyser stack size and service life must be considered when estimating the cost of hydrogen. For instance, PEM, alkaline, and solid oxide electrolyser stacks are expected to need replaced every 11, 9, and 7 years respectively, over a 30-year technology technical lifetime [33].

Another consideration for electrolysis scale-up is the required supply chain for the materials of manufacturing, including costly catalysts which are mainly made of noble metals such as iridium (Ir) and platinum (Pt). Details of the catalysts required for different types of electrolysers can be found in [34]. About 4.1% of the full economic cost is associated with the precious metallic catalysts in the cost breakdown for a 1 MW PEM electrolyser [34]. Assuming \$1500/kW of CapEx [35], the catalyst costs \$61,500 for a 1 MW PEM unit. In terms of weight, approximately 1.3 g/kW of iridium and 0.5 g/kW of platinum are currently required in an optimised PEM electrolyser [34]. This is in addition to the porous titanium that is also essential in PEM units. In another example, M. Chatenet et al. [36] in their review of water electrolysis technologies concluded that – assuming PEM is used for all the electrolysis – ca. 2.7 times the current annual rate of global Ir production is required to meet the 2030 target of the European Union alone (i.e., 40 GW of electrolysers). Anion exchange membrane (AEM) electrolysers are a rather new topic of research and have the potential to mitigate precious metal consumption while maintaining the electrolysis efficiency similar to the PEM units [37].

Currently, the most robust electrolyzers are alkaline, which use cost-effective nickel and potassium oxide in their catalytic construction, but are less flexible during fluctuating power flow and perhaps less energy efficient [38]. On the other hand, solid oxide electrolyser cells (SOECs) have been reported to reach energy efficiencies of >90% [39], but they are still under development and pending large-scale commercialisation. Overall, there is a trade-off between cost, durability, and efficiency when it comes to selecting the type of electrolyser. The latest reports on the raw material requirement for various electrolyser technologies are summarised in Table 1.

Despite the challenges mentioned above, and as the focus of this review is to establish whether H₂ for domestic heating would indeed neutralise the GHG emissions from this highly emitting sector, we will continue with the assumption that the production of sufficient green H₂ is realised.

3. Transmission and distribution

To follow the roadmap to decarbonisation of heating, e.g., by 2030 in the case of the United Kingdom, short-term and long-term strategies have been presented by the British Government. One of the short-term strategies is to blend green H₂ with the NG (HENG) in existing pipelines with minimum upgrade of infrastructure.

In view of this, this section aims to collect information on the readiness of the current infrastructure and to accurately describe the potential problems that may be associated with H₂ blends in NG pipelines such as material/equipment compatibility and leakage. This includes pipeline material degradation by H₂-induced phenomena (i.e., H₂ embrittlement and H₂ attack), unique thermodynamic behaviour of H₂, and H₂ compressibility which differs from NG. Furthermore, available literature on the matter is collected and presented to provide points of reference for individual thermodynamic problems faced in such systems. Finally, the potential effect of H₂ addition on the environment and easy-to-grasp economics are touched upon.

3.1. Current infrastructure readiness

A large percentage of households, especially in developed countries, are connected to the NG grid for their heating demands. For instance, about 85% and 50% of households in the United Kingdom (excluding Northern Ireland) [43] and the United States [44], respectively, use NG grid and boilers for heating purposes. Therefore, considering the broadness of the NG grid, its compatibility with the addition of H₂ is of great financial and temporal importance.

To this end, Material Durability and Integrity Management Institute of Gas Technology (IGT, 1972) stated that up to 20% by volume of H₂ could be added to the existing NG pipeline infrastructure in the United States with no modifications necessary [45,46].

H₂ can react with metals and form hydrides. This is the main principle for H₂ storage in solids. The same principle can potentially damage the metallic infrastructure in the current gas networks. H₂ penetrates the metal by two known mechanisms, namely hydrogen embrittlement and hydrogen attack. It should be noted that these phenomena are still under

research to find a more accurate description of the mechanisms [47,48]. Unfortunately, these two phenomena are mistakenly used interchangeably on occasion. To this end and for further clarification, the differences and similarities between these mechanisms are discussed below:

1. H₂ embrittlement:
 - a. It usually occurs at room temperature under high pressures.
2. In this phenomenon, the H₂ molecule breaks into H atoms by adsorption/desorption process on the wall, after which the atoms easily penetrate the defects of the metal lattice structure [49]. Then, migration of H atoms and/or formation of H₂ molecule within the lattice contribute to defect propagation and eventually (and perhaps under small external stress) lead to mechanical failure of the material [50]. H₂ attack:
 - a. It occurs at high temperatures (>204 °C [51]), and hence is also referred to as a high-temperature or hot H₂ attack.
 - b. It is fundamentally different from H₂ embrittlement [48], H₂ attack is instigated through the same adsorption/desorption process on the metallic surface.
 - c. In this case, H atoms can cause damage by bonding to other elements like carbon, forming heavier molecules such as CH₄ within the lattice structure or on the metal-gas interface. This is common in steels because of their carbon content. Dissociation of C from steel, decarburisation, can result in fissures and cracking compromising the mechanical durability of steel pipelines when used for transporting H₂ or HENG.

United Kingdom National Transmission System (NTS) comprises over 7600 km of steel gas pipelines [52] and usually operates at pressures >40 barg [53]. Distribution networks in the United Kingdom are over 284,000 km long [54] and operate at a pressure range of 0.03–7 barg [55]. The pipes transporting gas at pressures of >2 barg, 0.075–2 barg, and < 0.075 barg are typically made of steel, steel or polyethylene or cast iron or ductile iron, and polyethylene or cast iron or ductile iron, respectively [47].

United States transmission pipelines operate with pressures of up to 139 barg in mostly medium-strength steel. Gas distribution is also primarily made from low-strength or medium-strength steel, typically API 5 L A, B, X42, and X46 [56]. The lower the strength of steel, the less susceptible a pipe is to H₂-induced degradation. Thus, the type of steel pipes used in the United States gas distribution networks are assumed to be capable of handling H₂. However, further analysis on a case-by-case basis should be conducted for a safe and reliable repurposing of the pipeline for H₂. The report provided by [56] states ductile iron, cast and wrought iron pipes, and copper, are resistant to H₂-induced degradation and can be safely used with normal NG distribution conditions. Additionally, polyethylene (PE) or polyvinylchloride (PVC) pipe materials are deemed safe for H₂. In the case of existing measuring modules for gas flow rate, a deviation of <4% after recalibration is considered acceptable for HENG with H₂ content of up to 50 vol%. Overall, the Integrity Management Program (IMP) is reported to require minor infrastructure changes with H₂ content of up to 50 vol% [56]. It should be noted that the volumetric ratio of the H₂ admixture translates into different energy ratios in the HENG blend depending on the pressure of the blend or the individual constituents. For example, at normal temperature (15 °C) and pressure (1 barg), 50% of H₂ accounts for only 22% of the energy content in the blend, and if compressed H₂ at 350 barg is mixed with NG at 80 barg would yield an almost linear correlation between vol% and energy percentage. This must be also considered when designing the HENG transmission/distribution platform.

Although the existing pipe materials in the United States are deemed safe for low concentrations of H₂, ensuring the fire and explosion safety of distribution networks likely requires even lower concentrations of H₂ and more frequent inspection.

The leakage report by IGT from ductile iron and steel distribution pipes carrying HENG states that most of the leakage occurs through

Table 1

Some of the raw materials required for various electrolysis technologies in the near future (by 2030). Adapted from [36,40,41].

| | Alkaline | PEM | AEM [41] | SOEC and PCC ^e * |
|------------------------------------|---------------------------------------|-------------------------------|-------------------------------|--|
| Metallic elements required [kg/MW] | Ni: 800 C-steel: 10,000 Al: 500 | Pt: 0.3 Ir: 0.7 Ti: 500 | Pt: 0.1 Ir: 0.3 Ti: 500 | Ni: 200 Zr: 40 La, Co: 20 Y: 5 S-steel: 10,000 |

* Proton conducting ceramic electrolyser [42].

joints and threads [56]. The leakage rate from these pipes containing H₂ is 3 times higher than pipes with NG [56]. Elastomeric seals and plastic distribution pipes are more prone to H₂ permeation leakage. Although, the leakage rate through the elastomeric seals is higher than plastic pipes since the surface area of pipes is considerably higher than seals, leakage through the pipe walls accounts for the majority of lost H₂ from plastic piping platforms [56]. It is estimated the permeation rate of H₂ from such platforms is about 4 to 5 times higher than what it is for NG [56]. As an example, using a HENG with 20 vol% H₂ in a polyethylene pipeline results in losses where 60 vol% and 40 vol% of the leaked gas are H₂ and NG, respectively [56]. Even though the leakage volume could be twice as much as for NG [56], assuming a competitive levelised cost of H₂ with NG, it still remains economically viable as the cost of energy lost is less due to the lower volumetric energy density of H₂. Overall, H₂ blends can reduce the net NG emissions due to leakage [56]. A case study carried out by Haines et al. [57] on the Dutch pipeline system estimates a gas leakage rate of 0.00005% with a 17 vol% H₂ blend.

The report commissioned by the UK Government assesses the risk of 100% H₂ in the grid [58]. It concludes that in order to adhere to safe practices with H₂ in the pipelines, several measures, including the instalment of excess flow valves and vents in enclosed spaces, must be taken. In addition to further inspection and tight testing of the piping, any cast iron component must be removed or replaced. These recommendations imply additional labour and equipment costs to be added to the CapEx and OpEx of a fully H₂ gas platform.

In general, these reports imply that H₂ leakage from plastic distribution pipelines seems to be no major concern, but leakage from plastic service lines may pose a hazard for confined spaces. Therefore, more data on the leakage rate from the existing piping must be obtained in order to make executive decisions on the use of H₂ or H₂ blends in the NG pipelines.

On the other hand, pressure-reduction stations coupling transmission and distribution networks imply the need for significant pressure modulation, and H₂ does not behave similarly to NG when its pressure fluctuates. H₂ along with helium and neon, also known as quantum gases, are the only three gases that have a negative Joule-Thomson coefficient (μ_{JT}) (see Eq. (1)) [59]. Joule-Thomson expansion refers to when a gas or a liquid at a higher pressure is adiabatically transferred to a lower pressure with no significant change in the kinetic energy. The μ_{JT} quantifies the temperature change produced as a result of this expansion; it is positive when cooling and negative when heating occurs during the expansion. Therefore, when working with H₂ in a pressure-reduction station, chilling equipment and measures should be carefully considered to ensure a safe operation [60]. As a good example, J. Li et al. [61] studied the variation of μ_{JT} as a function of temperature and pressure for HENG with a hydrogen content of up to 30 vol%, showing a positive value even at a pressure of up to 100 barg. Fig. 1 shows the results obtained by extrapolating the linear trend reported by J. Li et al. [61]. As seen here, for both low-pressure and high-pressure conditions, 70 vol% of H₂ appears as the limit for a positive μ_{JT} value. Therefore, compression of HENG with 70 vol% of H₂ should not pose serious issues in pressure-reduction stations and the existing equipment could be utilised.

$$\mu_{JT} = \left(\frac{\partial T}{\partial P} \right)_H \quad (1)$$

Another important thermophysical characteristic of H₂ or HENG that needs to be considered is the compressibility factor (Z) (see Eq. (2)). This parameter shows how much a gas can be compressed at a given temperature and pressure, and it determines how gas behaviour deviates from ideal gas with a Z value of 1. However, the compressibility of H₂ even at low pressures is >1 , implying the demand for upgraded measuring and compressing equipment. This must be taken into account when adding H₂ to NG pipelines. For instance, S. Kuczyński et al. [62] show the correlation between the pipeline diameter and H₂ content in

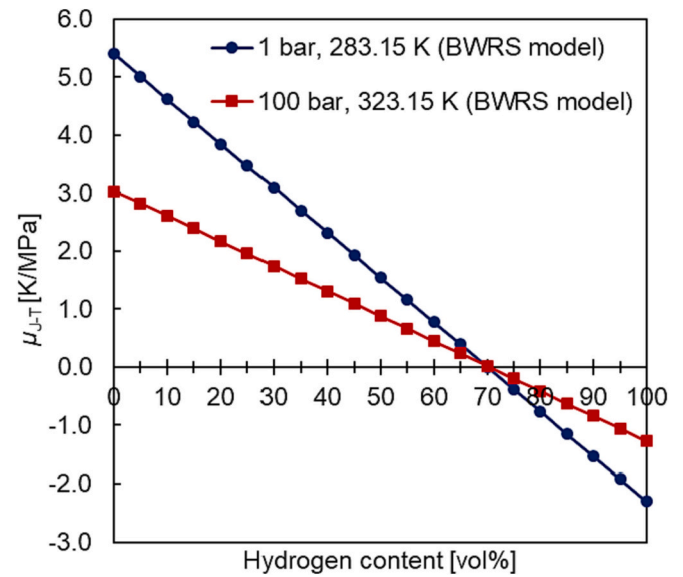


Fig. 1. Joule-Thomson coefficient of HENG with various H₂ content. This figure is adapted by extrapolation of the data from [61].

HENG to maintain a similar flow rate as NG. This further implies the potential need for upgrading the pipe sizes if H₂ or HENG blends are to carry an equal amount of energy as pure NG.

$$Z = \frac{P}{\rho R_{\text{specific}} T} \quad (2)$$

where, P , ρ , and T are the pressure, density, and temperature of the gas, respectively; R_{specific} is obtained by dividing the gas constant R by the molar mass of the gas (M).

Additionally, the change in the density and LHV of HNEG blends compared with NG would dictate the required pressure at compression stations and the diameter of the pipeline. For instance, S. Kuczyński et al. [62] show the correlation between the pipeline diameter and H₂ content in HENG to maintain a similar flow rate as NG. This further implies the potential need for upgrading the pipe sizes if H₂ or HENG blends are to carry an equal amount of energy as pure NG.

3.2. Environmental and economic cost of H₂ in pipelines

H₂ provides a means of storing RE at peak production and consequently mitigating the amount of curtailed energy. To convert H₂ produced in typically remote locations – where RE farms are located – back to power, a reliable transmission and distribution platform is required. There are multiple mainstream ways to store and transport H₂:

1. As the compressed gas;
2. Liquefaction;
3. Hydrogen-rich chemicals and synthetic fuels.

Compressing gas is energy-intensive and requires lightweight vessels that can withstand very high pressures, typically around 700 barg. As mentioned in the previous section, another alternative is to introduce the generated green H₂ into the existing NG pipeline. However, since H₂ is the lightest and smallest molecule known, the risk of leakage from the pipeline walls and conjunction points is higher than NG.

The latest report commissioned by the European Union [63] states that H₂ leakage from the grid has indirect effects on global warming by reducing species such as OH⁻ radicals and hence increases the lifetime of GHG gases like CH₄ [64]. The leakage also amplifies the global warming potential (GWP) of atmospheric water vapour, which then acts in a positive feedback loop to keep the heat from escaping from the earth,

more than doubling the warming effect of CO₂ increase alone [65]. Atmospheric water vapour is the only GHG that is condensable and hence, its concentration changes with the atmosphere temperature [65]. Overall, H₂ is estimated to have a GWP100 (over a 100-year period) of 11 ± 5 [66]. Therefore, the reduction of GHGs by capture and emission abatement is on par with large-scale detection and termination of leakage points. Some research suggests that a 10% leak in hydrogen could revert the environmental benefits of this carbon-free energy carrier [67]. This implies a need for rapid research on mobile detectors for deployment in large areas, and also for reducing the chance of CH₄ and other GHG emissions. The former involves effective equipment for rapid and large-scale monitoring of the mechanical infrastructure transmitting H₂. This could be achieved by employing drones equipped with sensors for non-destructive rapid inspection of the pipelines. Ultrasonic fault detectors could be an option for this purpose [68,69].

Current estimates predict a loss range of 1–4% for compressed H₂, up to 10–20% for liquefied H₂, and 0.7–1% for gas pipelines [63]. Though using H₂-ready pipelines for transportation results in the least leakage compared with compressed gas and liquified H₂, H₂-ready pipelines, require upfront investment for upgrading.

As discussed above, there are several uncertainties regarding the effect of H₂ on global warming and its mitigation measures. Some recommendations are:

- (i) to enhance the leakage detection technologies;
- (ii) enhance atmospheric sinks for H₂ (i.e., soil bacteria [70]);
- (iii) continue efforts to reduce methane emissions [71,72].

It is noteworthy to mention that contradictory to CH₄, optical detecting of H₂ is quite challenging as it does not show on the IR spectrum. One of the few available methods is Raman scattering, but this requires high-power illumination and sensitive detectors [63].

Finally, using H₂ in pipelines may take not only environmental tolls but certainly economic as well. F. Bainier and R. Kruz [73] investigated different volume fractions of H₂ in international NG pipelines. Assuming the infrastructure is already H₂-ready, they concluded that it takes 4 times the energy to transport the same energy quanta *via* H₂ than NG. It is estimated that the capital cost of H₂ pipelines is at least 10% more costly than NG pipelines [74]; however, this study lacks citations on the estimated price of the pipeline. In their report, Florisson et al. [75] also estimated that modifying the existing integrity management practices may result in a 10% additional cost when H₂ blends are to replace NG in the pipelines, irrespective of the percentage of H₂.

4. Combustion

In this section, the end-use of H₂ for heating is discussed in combustion terms. This includes the effect of H₂ loading in the fuel, air mixture by considering the equivalence ratio, and flame type on the efficiency and emissions of the boiler. In general, NG is composed of >95 mol% (or vol% if assumed ideal) CH₄ on average [76], and we assume NG is 100% CH₄ when discussing the chemistry of reactions.

4.1. Hydrogen content

Theoretically, increasing H₂ or H in the fuel reduces the environmental impact factor (EIF) of the fuel [77]. However, the H₂ percentage in the fuel also influences the combustion efficiency, NO_x, C_xH_y, and CO emissions. In general, to quantify the combustion energy, the Wobbe index (I_W) indicates the interchangeability of fuel gases by the ratio of the energy content of fuel (high heating value, HHV) to the specific gravity of the fuel (see Eq. (3)). The Wobbe index enables us to compare the combustion energy output for different composition of fuels in a unit (e.g., burner, cooker, boiler, etc) with the identical outlet pressure and valve (i.e., inlet nozzle to the combustion zone) settings. Thermophysical properties of NG and H₂ are summarised in Table 2, and Fig. 2 shows the

Table 2
Thermophysical properties of NG and H₂ [78–80].

| Property | NG | Hydrogen |
|--|----------|----------|
| Density (gaseous at STP) [kg/m ³] | 0.89 | 0.09 |
| Lower heating value [MJ/kg] | 46.9 | 120.0 |
| Higher heating value [MJ/kg] | 52.2 | 141.8 |
| Ignition limits [vol%] | 4.4–15.0 | 4.0–76.0 |
| Autoignition temperature [°C] | 540 | 585 |
| Minimum ignition energy [mJ] | 0.29 | 0.017 |
| Stoichiometric laminar burning velocity (LBV) [cm/s] | 41 | 237 |

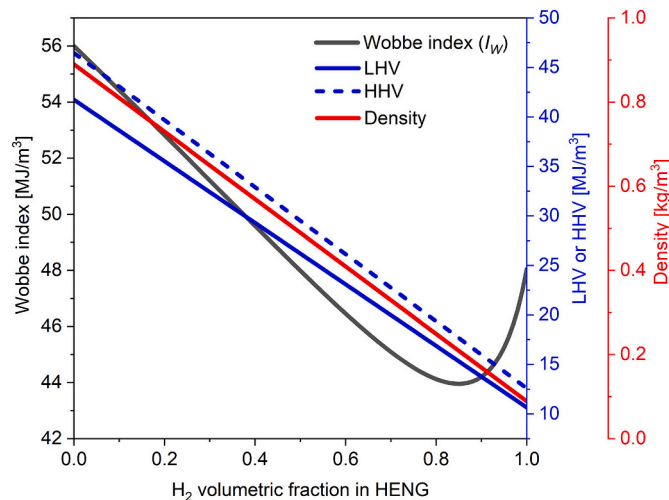


Fig. 2. HENG fuel energy indicators as a function of H₂ content.

dependence of combustion energy of the HENG on the ratio of H₂ content by volume. It can be seen that the addition of 20% H₂ to NG reduces the output energy of combustion by 5% using the identical pressure and valve settings. Replacing the NG with H₂ entirely results in a 10% reduction of combustion energy if the same inlet valve and pressure are used. However, with only 13% of H₂ in NG, the inlet valves are recommended to be upgraded which can then be recalibrated to to compensate for the output energy loss when 100% of H₂ is used instead of HENG.

$$I_W = \frac{HHV}{\sqrt{\frac{\rho_{f,STP}}{\rho_{air,STP}}}} \quad (3)$$

where, ρ represents the density and subscript STP implies the property to the standard temperature (0 °C) and pressure (1 atm).

4.2. Equivalence ratio and laminar burning velocity

Another combustion parameter that influences the quality of combustion and consequent emissions is the equivalence ratio (ϕ) (see Eq. (4)). 2% to 3% of oxygen and a small number of combustibles, 10–50 ppm, in the flue gas indicates an ideal operation for most systems [81]. A modern condensing boiler shows the optimal operation at $0.77 \leq \phi \leq 0.83$ [82]. If the optimal equivalence ratio for NG is assumed at 0.8, according to the shift of equivalence studied by H. de Vries et al. [83], optimal combustion of 100% H₂ would require an equivalence ratio of ca. 0.6.

$$\phi = \frac{\left(\frac{A}{F}\right)_{stoic}}{\left(\frac{A}{F}\right)_{stoic}} = \frac{E}{A} \quad (4)$$

In some cases, the air-to-fuel ratio (λ) is used when discussing

combustion. It is defined as Eq. (5).

$$\lambda = \frac{\frac{A}{F}}{\left(\frac{A}{F}\right)_{stoic}} = \frac{1}{\phi} \quad (5)$$

where, A and F are the volumetric flow rate of air and fuel, respectively. Subscript *stoic* stands for stoichiometric conditions.

Altogether, the equivalence ratio, smaller molecular weight, and fast diffusivity of H_2 compared with NG, affect the laminar burning velocity (LBV) which is the velocity of gas entering the combustion zone which further leads to how safe and noisy the nozzle is for H_2 or HENG jet flows. To eliminate the flashback hazard, the velocity of gas leaving the nozzle should be kept above the LBV at all times. Thus, LBV is an important parameter when selecting the diameter of the nozzle. Due to the high LBV of pure H_2 , flashback occurrence is more probable than many common fuels. C. Dong et al. [84] experimentally measured the LBV of HENG blends with varying H_2 content and equivalence ratio, which shows that 100% H_2 burns the fastest; The maximum LBV obtained for H_2 /air and NG/air mixtures are 2.933 and 0.374 m/s, respectively. This implies that with an identical combustor nozzle size, more H_2 is needed to be injected to increase the outflow velocity above the respective LBV, or the nozzle diameter should be smaller to increase the velocity above LBV for safety. Another phenomenon in combustion is backfire which is when the flame enters the nozzle and burns there with a whistling noise. ISO 13686:2013 designates the limit at 23 vol% of H_2 for burning HENG in the existing NG boiler where the limit for avoiding backfire is 35 vol% of H_2 [85] [86].

4.3. Flame type

Flame type is another influential parameter that should be considered in the design and effectiveness of H_2 -ready or H_2 -only boilers. Generally, there are two types of flames:

- (i) **Premixed** flame, where fuel and oxidiser are mixed before ignition;
- (ii) **Diffusion** flame, where fuel and oxidiser enter the combustion zone separately.

Both types have advantages and disadvantages which should be noted before designing an efficient burner. For instance, flame temperature, gas pressure, and emissions could be affected. Current gas boilers or cookers run on premixed flames. According to the study by M. Gazzani et al. [87], premixed H_2 combustion results in higher electric efficiency in an H_2 -fuelled steam turbine. A higher heat transfer rate in premixed flames was also reported by H. S. Zhen et al. [88]. In engines, premixed combustors offer leaner combustion with high efficacy and low NO_x emissions (e.g., dry low NO_x) [89]. However, premixed combustors present the risk of knock phenomenon and auto-ignition issues of the fuel/air mixture which can reduce the efficiency of the engine. Due to the higher combustibility of H_2 , such problems are more probable when using H_2 as the fuel in the premixed combustors [90]. Overall, there is still no consensus on the optimal design of the H_2 combustors as it also varies by application and required flame temperature.

CO and C_xH_y emissions may increase when blending NG with H_2 , as stated by [91,92]. Some studies mention negligible increases in these emissions. X. Zhan et al. [93] showed a decreasing trend for both CO and NO_x emissions using premixed HENG for a water heater with low heating loads (0.7–2.3 kW). However, premixed H_2 with air may pose hazards as H_2 is more combustible in a broader concentration range in the air (4.0–76.0 vol%). On the other hand, the efficiency and emissions of diffusion flame may differ significantly which could be a decisive factor in the design.

To sum up, the studies conducted on HENG [87–89] suggest that premixed flame leads to a more complete combustion and higher heat

transfer rate than diffusion flame. However, when transitioning to H_2 -only boilers, this has to be revisited with more experimental research.

4.4. Efficiency and emissions

Typically, the efficiency and emissions in a combustion device are correlated. Therefore, in this section, available reports on boilers operating on HENG or H_2 are summarised.

First, it is best to define efficiencies that are involved in such systems. Efficiency in boilers is usually rated with three definitions [94]:

- (i) **Combustion efficiency** (η_c) indicates how well the fuel is burnt and measures how much of it exits the burner unburnt. This is calculated using Eq. (8);

$$\eta_c = \frac{(H_2 \text{ mass flow rate}) - (\text{mass fraction of unburned } H_2 \text{ in the exhaust})}{H_2 \text{ mass flow rate}} \quad (8)$$

- (ii) **Thermal efficiency** indicates how well the heat exchanger transfers heat from combustion to water;
- (iii) **Overall efficiency** indicates how well the boiler converts the heat input from the fuel to the water.

From the aforementioned definitions, it is valid to expect the design of the whole system including the nozzle, burner, heat exchangers and heat exchanging mechanisms to affect the efficiency and emissions of the system. Hence, not only the combustion, but the balance of plant also matters greatly when discussing the efficiency and emission of boilers. GHG species that may be involved in the combustion of H_2 or HENG are listed in Table 3 to provide a better outlook in the following discussion on emissions.

Burning H_2 or HENG blends is primarily aimed at reducing CO_2 emissions associated with NG-fired boilers. To this end, F. Schiro et al. [103] conducted a case study on a condensing boiler fuelled with HENG. CO_2 emissions of HENG with different H_2 fractions are shown in Table 4. As seen here, the addition of H_2 at 20–80 vol% reduces CO_2 emissions by 7.3–55% in a condensing boiler. This analysis, however, does not consider the change required in the design of the boiler and it is purely mathematical. Overall, the reduction in CO_2 emissions even at a 7.3% rate on a million-ton scale would be a major milestone achieved on the path to decarbonisation. To give a sense of scale, an increase of 0.9% in energy-related CO_2 emissions in 2022 translates into 321 Mt. of which

Table 3
Global warming potential of GHGs [95].

| Species | Chemical formula | Lifetime | Global Warming Potential (Time horizon) | | |
|---------------------------------|------------------|---------------------|---|--------------------|-----------|
| | | | 20 years | 100 years | 500 years |
| Carbon dioxide | CO_2 | 5–200 yr [96] | 1 | 1 | 1 |
| Carbon monoxide | CO | 1–2 mos [98] | 2.8–10 | 1–3 | 0.3–1 |
| Hydrogen | H_2 | 2.4 yr [97] | 40.1 ± 24.1 [99] | 11 ± 5 [66] | – |
| Methane | CH_4 | 12 ± 3 yr | 56 | 21 | 6.5 |
| Nitrous oxide | N_2O | 120 yr | 280 | 310 | 170 |
| Nitrogen oxides (oxide/dioxide) | NO_x | variable (40–54 h)* | 30–33 [101] | 7–10 [101] | – |
| Water vapour | H_2O | – | – | –0.001–0.0005 [15] | – |

* NO_x lifetime is highly dependent on seasonal changes [102].

Table 4

CO₂ emission from a condensing boiler fuelled by HENG. Data adapted from [103].

| Fuel | CO ₂ emissions [g/MJ] |
|----------------------------|----------------------------------|
| NG | 54.6 |
| HENG (20% H ₂) | 50.6 |
| HENG (40% H ₂) | 45.4 |
| HENG (60% H ₂) | 37.2 |
| HENG (80% H ₂) | 24.5 |
| H ₂ | 0 |

60 Mt. is associated with heating and cooling [104].

Even though effective in CO₂ reduction, burning H₂ or HENG may result in the emission of other GHG species such as NO_x. Table 5 summarises the typical emission equivalent of available heating technologies according to the 2004 report by the World Energy Council [100]. Comparing the values between Tables 4 and 5, one can see that even for a conventional condensing boiler fuelled by NG, 27.2% (or 20.4 gCO₂eq/MJ) of the emissions are non-CO₂. Therefore, further emission investigation is needed to ensure the effectiveness of H₂ or HENG boilers in GHG reduction.

NO_x is another common emission in the combustion of fuels with air, especially in the case of H₂. Typically, higher NO_x formation with higher H₂ content is attributed to the presence of higher quantities of OH, H and O radicals leading to thermal NO formation [106]. Moreover, H₂ has a very high LHV and burns with a higher stoichiometric adiabatic flame temperature (2182 °C) than NG (1937 °C) [107]. This high temperature can potentially provide energy for breaking the stable triple bond of N₂ in air resulting in the formation of NO [108]. In light of this challenge, some studies have tried to experimentally evaluate the NO_x formation against the efficiency of boilers. For instance, in their study, T. Wang et al. [109] report the experimental results of a 180 t/h steam boiler fuelled with HENG (0–100% H₂). They concluded that there is a trade-off between the thermal efficiency of the burner and the NO_x emissions, especially with unchanged valve settings. G. L. Basso et al. [94] also concluded that the water dew point temperature in the exhaust gas decreases if the mixture is lean leading to lower condensation efficiency in the condensing boilers. The latest case study on domestic H₂ boilers by S. Gersen et al. [85] specifically investigates NO_x emissions from two retrofitted NG units that are H₂-ready with a power range of up to 23 kW. Their results show no major concern regarding NO_x using properly tuned and optimised H₂-ready boilers. However, there are no details on the optimisation method, perhaps due to confidentiality. Additionally, since domestic boilers could be rated up to 42 kW of heating power [110], the actual NO_x emissions for higher heating loads must be also measured. Finally, other studies also claim that a similar or higher combustion efficiency was obtained in boilers fuelled with HENG containing 25 vol% [111] and 30 vol% [112] of H₂, with no NO_x addition.

Other than CO₂ and NO_x, combustion of NG and H₂, either blended or individual, may result in some side reactions leading to unburnt hydrocarbons (UHCs) or the formation of new C_xH_y species that can potentially amplify the greenhouse effect. It should be noted that UHC refers to the exhausted feed fuel (e.g., CH₄) and C_xH_y refers to the other hydrocarbon byproducts such as CH and C₂H₃ which could be by-produced during the combustion of CH₄ [113]. This is more probable

Table 5

Equivalent CO₂ emissions (including all GHG emissions such as CO and CH₄ and based on GWPs) from various heating technologies. Data adapted from [105].

| Heating means – Energy source | Equivalent CO ₂ emissions [g/MJ] |
|--------------------------------------|---|
| Electricity – NG | 150.0 |
| Boiler – NG | 83.3 |
| Condensing boiler – NG | 75.0 |
| Ground source heat pump – NG | 36.1 |
| Ground source heat pump – RE (solar) | 2.8 |

when using HENG blends as more H atoms are available for reaction. Y. Zhao et al. [114] studied emissions from a HENG-fuelled condensing boiler; they concluded that UHC emissions remained constant or decreased as the H₂ was increased to 20 vol%. This study, however, does not analyse H₂ content of >20 vol%. Another case study in Ontario, Canada, showed the reduction of GHG emissions, mainly comprised of CH₄, using HENG instead of NG [115]; the addition of 5 vol% H₂ to NG pipelines reportedly reduced CH₄ emissions, however small (by 0.4% excluding NO_x). On the other hand, M.S. Boulahlib et al. [92] report an increasing trend in C_xH_y emissions when experimentally studying the operation of a domestic boiler fuelled with HENG containing up to 45 vol% of H₂. However, their experiments involve highly fuel-rich combustion with 2 ≤ φ ≤ 4. Overall, there are reports on the increase of UHC and/or C_xH_y emissions when using HENG with <50 vol% of H₂, and this should be further tested experimentally if HENG is going to replace NG in the short term. If proven consistent, mitigation methods must be identified.

Another species involved in HENG combustion is CO, a lethally toxic GHG. The conversation around the formation of this species is also not very unified. However, the same studies on UHC and/or C_xH_y emissions concluded an unchanged or increasing trend in CO when burning HENG [92,114]. This warrants further investigation on CO emissions from boilers fuelled by HENG.

N₂O and NH₃ are also some of the known species that could be produced during the combustion of fuels with air. However, the study by Y. Zhao et al. [114] on a HENG-fuelled condensing boiler shows negligible change in NH₃ and N₂O emissions. The reduction in N₂O emission by using H₂ as aviation fuel was again mentioned in the work of P. Gunasekar et al. [116].

Additionally, reports on the addition of H₂ to CNG (HCNG) for engines also imply a positive outcome in terms of efficiency and emissions. For instance, J. R. Anstrom and K. Collier [46] have collated the data on HCNG in engines. They concluded that up to 20 vol% of H₂ is attainable with no engine retuning required. Adding H₂ to the mix also improves the quality of combustion by mitigating the amount of unburnt CH₄ in the exhaust, or fugitive methane tailpipe emissions, by providing an abundance of hydroxyl ions to reduce methane. Moreover, adding H₂ by 30–50 vol% has been shown to significantly reduce NO_x emissions and improve engine efficiency compared with gasoline, CNG, or Hythane® (i.e., HCI trademarked blend of 20% hydrogen with NG to match gasoline flame speed).

Altogether, other than combustion conditions, the emissions from a boiler are also dependent on other parameters such as water flow rate in the heating system [114]. It is believed that more attention should be paid to CO and C_xH_y, rather than NO_x, emissions when discussing HENG blends. However, these emissions will be eliminated once pure H₂ is ready to burn in boilers as full H₂ replacement and complete abatement of GHG emissions from heating is the ultimate goal of the hydrogen-for-heating scheme. So far, reports of available case studies on H₂ boilers have demonstrated reasonable CO₂ and NO_x reduction and a comparable thermal operation with current NG boilers. It is worth noting that combusting 100% H₂ may offer a higher condensation efficiency in a condensing boiler, as the condensable water mass increases with H₂ content in the fuel [94,117]. When burned at a similar equivalence ratio with NG, more condensable water mass will contribute to utilising HHV of pure H₂ in condensing boilers which theoretically can lead to higher gravimetric energy efficiency of the boiler.

5. Conclusions

In line with climate actions around the globe, many nations have targeted to at least partially decarbonise heat by 2030 [118]. To achieve this rapidly in such a short time, utilising the existing infrastructure may present a fast and easy solution by introducing green H₂ to the NG grid. However, financial and technological resources must be allocated carefully to avoid the same long-term environmental impacts of fossil

fuels. Overall, during the operation of pure H₂ pipelines, leakage may lead to more environmental risks rather than economic ones. H₂ boilers could effectively decarbonise domestic heating, but the discrepancy between the current and required production rate of renewable electricity and green H₂, and the compatibility of infrastructure create most of the associated risks.

Findings:

1. The possibility of sustainable water resourcing for green H₂ is strongly dependent on the sizing, siting, and service life of electrolysis plants, as well as post-purification hard water treatment plans; more data is required.
2. HENG with H₂ content of up to 20–23 vol% is theoretically attainable using the existing boilers and infrastructure. Up to 50 vol% of H₂ will require minor changes to the transmission/distribution and measuring equipment across the supply chain. Above 50 vol%, however, major infrastructure change is required. Safety and compatibility of the current infrastructure are highly compromised with H₂ content of over 70 vol%.
3. Replacing NG with 100% H₂ demands additional CapEx and OpEx. This is mostly associated with transmission pipelines, pressure reduction stations, and safety measures in distribution systems. The integrity of transmission pipelines against 100%H₂-induced degradation with pressure above 139 barg is uncertain and therefore, requires further experimental research.
4. Leakage of H₂ from the pipelines does not pose a major economic risk, but its adverse environmental impact could weaken the GHG reduction potential of the H₂-to-heating platform.
5. CO₂ is certainly reduced by adding H₂ to NG. NO_x mitigation is achievable and the techniques are known. However, CO and C_xH_y, and UHC emissions require more experimental validation as some reports have found an increasing trend in such species when burning HENG.
6. Although we believe that the state-of-the-art boiler technology is capable of addressing suspected issues regarding the emission and efficiency of H₂ boilers, manufacturers are highly encouraged to publish their test results regarding the emissions of their H₂-ready and/or H₂-only boilers. To the best of our knowledge, there is very limited information available from the industrial players in this sector.
7. In order to hasten the transition and mitigate risks, other heating alternatives such as heat pumps and perhaps new disruptive technologies, active or passive, must be pursued in parallel.

CRedit authorship contribution statement

Arash Badakhsh: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Sivapriya Mothilal Bhagavathy:** Writing – review & editing, Resources, Project administration, Funding acquisition.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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Data availability

Data will be made available on request.

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