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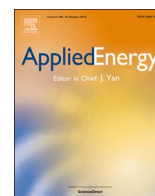
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The impact of natural gas/hydrogen mixtures on the performance of end-use equipment: Interchangeability analysis for domestic appliances



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HIGHLIGHTS

- Quantifies changes in performance for domestic appliances upon hydrogen admixture.
- Interchangeability analysis using calculated burning velocities and Wobbe Index.
- Calculated maximum hydrogen fractions verified by flashback measurements.
- Fuel-rich premixed appliances are critical for flashback.
- Composition of the gas to which hydrogen is added determines the maximum admixture.

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ABSTRACT

The addition of hydrogen derived from renewable power to the natural gas network is being promoted as a viable means of storing excess wind and solar energy. However, the changes in combustion properties of the natural gas upon hydrogen addition can impact the performance of the end-use equipment connected to the gas grid. We assess the changes in safety and fitness for purpose of domestic natural gas appliances when supplied with natural gas/hydrogen mixtures. Upon hydrogen addition, the fitness-for-purpose limits are governed by changes in thermal input caused by changes in Wobbe Index, while the changes in risk of flashback are used to quantify the safety aspects. A method is introduced to assess changes in the propensity for flashback, using the computed laminar burning velocity and accounting for changes in equivalence ratio caused by the variations in fuel composition. The computations are seen to reflect the experimental behavior of Bunsen flames regarding flashback upon hydrogen addition to natural gas accurately. Comparing the changes in Wobbe Index and variations in burning velocity with those experienced by domestic appliances subjected to the range of natural gases ordinarily distributed to the end user provides an unambiguous and internally consistent method to assess changes in essential performance upon hydrogen addition. Thus, limits on hydrogen addition can be derived to maintain the current level of safety and fitness for purpose without the necessity of large-scale appliance testing. The results show that the maximum fraction of hydrogen in natural gas that maintains appliance performance depends on the composition of the natural gas to which the hydrogen is added. For fuel-rich premixed appliances, e.g., cooking burners, the maximum hydrogen admixture is seen to be limited by flashback, while loss of thermal input determines the maximum hydrogen fraction in modern lean-premixed appliances. The method is illustrated using a fictitious distribution band, but it can be applied to any regional or national situation. The method presented can be used to define efficient strategic roadmaps and provides essential knowledge for grid management assessments, aimed at introducing hydrogen into the natural gas infrastructure while maintaining performance for the end user.

1. Introduction

The drive towards sustainability and energy independence is urging international policy to reduce CO₂ emissions and the dependence on

fossil fuels. Improving energy efficiency and increasing the capacity of wind and solar power generation is growing rapidly in several European countries. A concrete example is that in 2011 in Germany it was decided to strongly promote energy efficiency (households,

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Nomenclature

A	volumetric flow rate of (dry) air (m^3/s)	W	volumetric flow rate divided by cross-sectional area of burner tube (cm/s)
B	thermal input, or thermal load (for domestic appliances in the order of 1–10 kW)	W_{min}	Wobbe Index (MJ/m^3 (25 °C, 0 °C))
d	relative density of fuel compared to air (–) (0 °C)	W_{max}	Wobbe Index at lower limit 48.17 MJ/m^3 of fictitious distribution band used
$\Delta\Phi$	shift in equivalence ratio (–)	W_{min}	Wobbe Index at upper limit 57.66 MJ/m^3 of fictitious distribution band used
Φ	equivalence ratio (–)	W_{min}	gas composition $\text{CH}_4 = 92.6 \text{ mol}\%$, $\text{N}_2 = 7.4 \text{ mol}\%$, having $W = W_{min}$
F	volumetric flow rate of fuel (m^3/s)	W_{max}	gas composition $\text{CH}_4 = 87 \text{ mol}\%$, $\text{C}_3\text{H}_8 = 13 \text{ mol}\%$, having $W = W_{max}$
H_s	higher heating value (MJ/m^3 (25 °C, 0 °C))	AGA	American Gas Association
L_d	molar stoichiometric air requirement (mol/mol)	CNG	compressed natural gas
L_{dV}	volumetric stoichiometric air requirement (m^3/m^3)	NG	natural gas
m^3	cubic meter at 101.325 kPa and 0 °C		
$n(\text{O}_2)$	number of O_2 molecules for complete combustion of 1 fuel molecule (mol/mol)		
p	pressure (kPa)		
S_L	laminar burning velocity (cm/s)	<i>Subscripts</i>	
T	temperature (°C)	<i>adj</i>	adjustment of appliance
T_{ad}	adiabatic flame temperature (K)	<i>s</i>	superior
ν_u	velocity of unburned mixture from burner exit (cm/s)	<i>stoich</i>	stoichiometric
$\nu_{u,f}$	velocity of unburned mixture at flashback, defined as	<i>w.c.</i>	worst case

industry, buildings) as well as renewable electricity generation [1], while also cutting down on nuclear energy [2]. Since wind and solar energies are ‘intermittent’, using the natural gas grid in countries with a well-developed gas infrastructure to act as a buffer is a common theme in policy literature, emphasizing the need for integrated approaches along the entire electricity and gas value chains [3–8]. An increasingly important role for hydrogen as an energy carrier has been foreseen [9,10], particularly in periods of overproduction of renewable electricity, as compared to the capacity of the power infrastructure [3,4,11,12]. Hydrogen can be produced with a maximum degree of sustainability through electrolysis (see [12,13], for example) using wind or solar energy as a power source and water as carbon-free source of fuel.

The conversion of electrical energy into gaseous chemical energy thus appears as a promising concept called “power-to-gas” [3,6,7,11–13]. While the primary step in this process is hydrogen production, in a potential second step methane can be produced from the combination of hydrogen and renewable carbon dioxide from, for instance, fermentation gas. While hydrogen and methane can both be used as transportation fuels, injection into the natural gas system is an obvious candidate for large-scale buffering of this excess renewable energy. In Germany, for instance, utilizing the natural gas network in this way is considered the only option for a significant amount of storage [4,11,14].

Recent studies (for example, [3–7,10,15]) have focused on various ‘system’ aspects of the natural gas/hydrogen system. Energetic and capacity aspects of transporting natural gas/hydrogen mixtures in natural gas pipelines have been studied [16–18]. Case studies have analyzed the technical requirements for ‘power-to-gas’ in Germany [4,7], and integration and economic aspects of coupling the electricity grid to the gas network using hydrogen in the UK [6,8] and in the province of Ontario [12]. The choice of the location of hydrogen injection points and the use of natural gas/hydrogen mixtures in different end-use applications (domestic, industrial, power generation) downstream of the injection points pose challenges to grid management. The issue of grid management is being addressed in several studies at present. Hydrogen injection at different nodes in a low pressure natural gas network has been modeled, showing the impact on pressure distribution and gas composition (usually denoted as gas ‘quality’) in the network [19]. Hydrogen injection has been implemented in a dynamic model of natural gas quality in a transport pipeline [20], while maintaining the load profile (energy demand), obtaining composition, flow

rate and pressure profiles. Hydrogen blending has been studied in a numerical simulation of a regional scale natural gas transmission system under steady-state condition with a newly developed model [21] showing the impact on gas quality. These network studies identify the fact that information on hydrogen fractions that gas-utilization equipment can tolerate is insufficient as yet. This marks an important knowledge gap for the assessment of gas grid management when considering the possibility of hydrogen injection. Despite this lack of information, these studies make explicit assumptions about the degree of hydrogen admixture that will not adversely impact the performance of end-use equipment. Various maximum hydrogen fractions have been assumed depending on the specific class of end-use equipment and local regulation, ranging up to 20% cited for domestic equipment [7].

However, to our knowledge there is very little evidence demonstrating that the adequate performance of the population of end-use equipment installed in the field is *guaranteed* for any degree of hydrogen admixture. While a substantial number of studies have been published on the impact of hydrogen/natural gas mixtures on the performance of automotive engines using compressed natural gas, CNG (see [22,23], and references therein), little attention has been paid to the general analysis of the impact on the performance of domestic equipment, the focus of the current study. The studies reported to date have been limited to fundamental combustion studies of phenomena related to the performance of domestic appliances [24,25] or to the laboratory investigation of some practical appliances [26].

Since domestic appliances are connected to the natural gas grid in very large numbers, and that the admixture of hydrogen can change the combustion properties of natural gas substantially, the lack of information on their performance when fueled with hydrogen/natural gas mixtures forms an essential uncertainty for grid management analyses. Despite preliminary considerations [10], clearly motivated and fundamentally justified hydrogen fractions for populations of domestic appliances are not available at present. A gas interchangeability method, which also explicitly addresses the specific combustion phenomena affected by hydrogen addition (i.e. flashback, see below), is needed that is applicable to any individual natural gas distribution situation, while avoiding the necessity of testing large numbers of locally installed appliances. The availability of such a method would bridge this critical knowledge gap.

In general, hydrogen addition to natural gas decreases the Wobbe Index and increases the so-called laminar burning velocity, S_L . As discussed in detail below, the Wobbe Index is directly related to the heat

input to domestic appliances, and is an essential requirement for the fitness for purpose from the perspective of the end user. The burning velocity is the property of the fuel that is intimately related to flame stability issues such as flashback [27]. Flashback can cause damage to the burner or flame extinction, which, in the absence of a flame safety device, can result in spillage of combustible mixture into the home. This safety risk is why flashback and its consequences must be avoided. The changes in burning velocity upon hydrogen addition to natural gas (NG) have been studied experimentally (for example, [24,25]), showing a substantial increase in burning velocity with increasing hydrogen fraction. These studies have been invaluable in verifying burning velocity computations [28], but did not analyze the possible consequences of the changes in burning velocity for domestic appliances. The laboratory investigation of domestic appliances [26] examined the performance of a small number of lean-premixed appliances with varying methane/hydrogen mixtures. While variations in performance and emissions were observed, no flashback was reported. As will be discussed below, the scope of that study was not representative of the installed base of appliances, nor of the range of natural gas compositions distributed.

Thus, while important information regarding fundamental changes in combustion phenomena and the performance of individual appliances has been obtained, the scope of studies performed to date does not allow conclusions regarding the maximum fraction of hydrogen that maintains the performance of domestic appliances installed in the field. In this study, we analyze and quantify the effects of hydrogen addition on the combustion performance of domestic equipment, regarding fitness for purpose and safety, particularly flashback, to permit conclusions for the entire population of installed appliances. Towards this end, we perform an interchangeability analysis based on the computation of burning velocities. The method presented below allows conclusions at a level of detail that has not yet been reported in the literature.

2. Hydrogen in the installed base of domestic appliances

As mentioned above, the situation that must be assessed when considering addition of hydrogen to an existing natural gas infrastructure is that of a large (roughly 190 million in the EU alone) and extremely diverse population of *installed* appliances. Since natural gas has been a major source of domestic energy for many decades, and that domestic appliances can easily have a natural lifetime of more than 15–20 years, the installed base of appliances reflects the large number of appliance types and designs developed during this span of time. In addition to the variety of types, such as central heating boilers, hot water heaters, cookers, ovens and decorative fires, there are distinctions in the type of combustion system. As will be seen below, particularly important is the wide range of primary fuel–air ratio in these appliances, varying from ‘fuel rich’ to ‘ultra lean’; these different types of appliance respond differently to hydrogen addition. Additionally, the individual appliances have each been subject to a different history of installation, adjustment and maintenance, resulting in appliances that, through proper adjustment and maintenance, can be robust towards changes in gas composition, or alternatively, through maladjustment and/or lack of maintenance, be at (or even beyond) the limit of acceptable performance. These issues have also been discussed in the context of variations in natural gas composition [29].

Another aspect that will be seen to be essential in the analysis of the population of domestic appliances is that the composition of the natural gas to which hydrogen is added is itself not constant. Each country, and even local regions within a country, has a variation in gas composition that will mitigate or aggravate the effects of hydrogen addition for end-use equipment.

Given this diversity in appliance population and natural gas composition, it will be clear that performing a few experiments testing the response of a limited number of domestic appliances when supplied with hydrogen-containing natural gases under laboratory conditions

(for example, as done in [26]) is insufficient to generalize conclusions to the installed base. Experimental determination of the degree to which the installed base of appliances can accept hydrogen-containing natural gases requires large-scale testing of representative samples of appliances, with a representative range of gas compositions, in the field. Such a large-scale study is logistically daunting and costly. Here we describe a different approach, which does not require large-scale appliance testing, but relies on the analysis of the characteristics of the range of natural gases that is deemed to be acceptable for the appliance population, i.e., a so-called interchangeability analysis.

3. Interchangeability analysis

Two gases can be considered interchangeable when substitution of one gas for another does not lead to significant deterioration in the performance of a given appliance. Clearly, quantifying ‘significant’ deterioration in performance can be subjective, with different countries having different criteria. Below we shall provide examples of an internally consistent method of setting limits on acceptable behavior. However, before proceeding it is important to emphasize that comparing the properties of gases can result in clear boundaries for gas composition, but at the same time provides no information about the performance of individual appliances.

Interchangeability parameters are usually defined for critical aspects of appliance performance. These can be subdivided in aspects for fitness for purpose and those for safety. The major aspect regarding the characterization of gaseous fuels for fitness for purpose is the thermal input to an appliance. Clearly, if the thermal input to an appliance will change significantly upon changing the fuel gas, then this should be known in advance. As can be easily shown, the thermal input to a domestic appliance is proportional to the ratio H_s/\sqrt{d} [30–33], where H_s is the higher heating value and d is the relative density of the fuel compared to air, comprising the well-known definition of the Wobbe Index, W [34,35]. Which variations in thermal input upon varying the fuel composition are deemed acceptable is often based on the traditional ranges of gas supply [36]. Regarding safety, a stable flame is a prerequisite for safe operation, since loss of flame stability can lead to escape of explosive mixture and/or excess CO emissions into the living space. Similarly, insufficient oxygen supply for a given fuel can lead to excess CO emissions. Thus, critical safety aspects that are generally included in interchangeability analyses are the flame stability issues flashback and flame lift, and incomplete combustion, specifically CO emission and soot formation [32,33,37].

While the relation between the thermal input and the Wobbe Index has been amply verified experimentally, the safety aspects of interchangeability have been thus far characterized empirically. While the AGA interchangeability method utilized Bunsen flames [32], other methods [33,37] assessed a set of appliances deemed representative for the end user exposed to a range of fuel compositions and observed the changes in the critical aspects of appliance performance. Relations were then derived to quantify the effects of fuel composition. The functional response of flame stability and incomplete combustion observed when varying the fuel composition, as well as the limits set on acceptable variations in performance, are governed by the set of appliances/burners used to derive the data, all of which are 40–70 years old. Recently, the accuracy of the AGA Lift Index has been examined by tests with 17 Chinese domestic gas cookers fueled by 11 test natural gases. The tests showed that the interchangeability limit values of this Index needed to be changed to reflect the behavior of the burners tested ([38], and references therein). However, the alterations in the limit values derived in those experiments are necessarily restricted to the set of appliances and gases used in the tests. This example illustrates the fact that the empirical methods traditionally used do not yield useful interchangeability results for situations in which newer and/or different types of appliance are in use than those originally chosen, and where possible effects of (a lack of) maintenance on appliance performance must be considered. We

note that one of these methods [37] has even been written into national law [39].

In the present study, an interchangeability approach is applied that is based on characterizing the changes in performance as a function of fuel composition using fundamental combustion parameters [40], rather than depending on the vagaries of the performance of the combinations of appliance design parameters. To be accurate, this approach also includes an important operational principle: in the absence of an active control system, the primary fuel–air ratio in an appliance changes with fuel composition [31]. The primary fuel–air ratio is the dominant property that changes when changing fuel, having large impact on flame stability and incomplete combustion, as will be demonstrated below for flashback. For each performance aspect, a fuel gas is characterized by a specific combustion parameter. For example, when characterizing flame stability such as flashback the laminar burning velocity S_L is the relevant combustion parameter [27,41–43]. Here, we describe the interchangeability method in detail and apply it to analyze the consequences of adding hydrogen to natural gas. While a number of the examples are taken from the natural gas distribution practice in the European Union, the results are perfectly general and, as will be seen below, straightforwardly applicable to any specific situation.

4. Properties related to end use: comparison with methane

The specific importance of hydrogen in the present context is that its occurrence in natural gas is negligible ($< 0.1\%$ [44,45]), while it has strongly different properties related to end use as compared to those of typical natural gases. Hydrogen also shows specific interaction with materials (metals, fitting materials) not manifest by natural gases in the entire infrastructure [10,46], which is outside the scope of the present study.

As methane, CH_4 , is the primary component of natural gases [47] it is instructive to compare several properties of CH_4 and H_2 that are relevant for end use, given in Table 1. In this work, superior calorific values (H_s) and relative densities (d) of mixtures are calculated applying the methods outlined in [34].¹ Further, in the table are given: $n(\text{O}_2)$, the number of oxygen molecules needed for the complete combustion of one fuel molecule, the molar stoichiometric air requirement L_b , the number of moles of air required for complete combustion of one mole of fuel, the volumetric stoichiometric air requirement L_{dv} , in m^3 air required for 1 m^3 fuel, and burning velocities (S_L) and adiabatic flame temperatures (T_{ad}), both at the stoichiometric fuel–air ratio. The fuel–air ratio is expressed as the equivalence ratio, Φ , that is defined as the actual fuel–air ratio divided by the stoichiometric fuel–air ratio; for a stoichiometric fuel–air mixture, $\Phi = 1$. In addition, the ratios of the properties listed for the two fuels are given in the table.

Given the large differences between properties shown in Table 1 for hydrogen and methane, substantial changes in these properties are to be expected from admixing H_2 to natural gas, with concomitant impact on the performance of domestic appliances, as will be described below.

Before discussing the specific aspects of appliance performance, with an eye towards the possible contribution of hydrogen admixture towards reducing the CO_2 burden of natural gas, we first consider the effect of admixing H_2 on the potential CO_2 emission reduction that can be obtained. For this purpose, we consider natural gas as pure methane, without loss of generality. While it is tempting to consider the CO_2 reduction as being directly related to the volume of methane replaced by hydrogen, and of course 100% replacement of natural gas by

¹ For the accuracy of calorific value calculations on a volumetric basis the methods given in Ref. [35] pose limitations on component fractions, which are exceeded here, thus [34] is used. In the table, the reference conditions (p , T) used for the parameters are (101.325 kPa, 25 °C) for thermodynamic quantities and (101.325 kPa, 0 °C) for volumes. For properties such as H_s and W , the combination of thermodynamic (25 °C) and volumetric reference temperatures (0 °C) is commonly referred to as (25, 0), and used here. The set of reference temperatures (15 °C, 15 °C) is also common practice [48,49].

Table 1
End-use properties.

Parameter	CH_4	H_2	Ratio (CH_4/H_2)
H_s (MJ/m^3)	39.82	12.75	3.1
d (0 °C) (–)	0.555	0.070	7.9
W (MJ/m^3)	53.45	48.34	1.1
$n(\text{O}_2)$ (mol/mol)	2	0.5	4
L_b (mol/mol)	9.5484	2.3871	4
L_{dv} (m^3/m^3)	9.5657	2.3845	4.012
S_L (cm/s)	36.7	203.9	0.180
T_{ad} (K)	2226	2381	0.935

As indicated in the text, H_s and W are taken at the reference conditions (25, 0), and S_L and T_{ad} are taken at $\Phi = 1$.

hydrogen means a 100% reduction in CO_2 emissions, it is important to recall that the utilization of natural gas in domestic appliances requires a given, constant, total heat input: for heating, for example, raising the temperature of water in a system of radiators requires a certain heat input, independent of the fuel composition. However, referring to Table 1, the heating value of the mixture (which varies linearly with admixture) decreases with hydrogen addition. Thus, more fuel (producing more CO_2) is required to achieve the same temperature rise in the radiator water. The net effect on CO_2 is shown in Fig. 1 for CH_4/H_2 mixtures. The CO_2 reduction is in fact equal to the reduction in the CH_4 contribution to the constant energy input. Obviously, the increase of the energy fraction delivered by H_2 shows the same functional dependence. Consequently, the use of more fuel substantially diminishes the impact of hydrogen addition to methane; at fractions below $\sim 30\%$ hydrogen, the CO_2 reduction is only 1/3 of the hydrogen fraction, tempering the expected impact of hydrogen addition to natural gas on CO_2 emissions.

5. Changes in thermal input with hydrogen addition

Whereas adding hydrogen itself tends to increase the risk of flashback in appliances, making consideration of this safety aspect inevitable, discussing the effects on thermal input is essential for assessing the impact of hydrogen admixture on the fitness for purpose of the fuel for the end user, and comprises a necessary element in the present interchangeability assessment. While examination of Table 1 shows that replacing 100% CH_4 by 100% H_2 will decrease W by a factor of 1.1, W does not simply linearly decrease with increasing hydrogen fraction. Upon admixing limited amounts of hydrogen, the Wobbe Index of the fuel decreases considerably faster.

As anticipated in Section 3, above, an important aspect for the end user of combustion equipment is the thermal input, B , which is simply the volumetric flow rate of fuel (F , in m^3/s) multiplied by the higher heating value, $B = F \times H_s$. The thermal load of an appliance determines the level of comfort provided by it, in terms of its ability to provide enough heat to heat a home or perform cooking tasks. Generally, when the gas supply pressure to domestic appliances is constant,² [31,41]

$$F \propto 1/\sqrt{d}; \quad (1)$$

recalling the definition of the Wobbe Index, W , given above, we see that $B \propto W$. The Wobbe Index is thus a convenient measure to compare the

² Calculations in [8] indicate that when the energy transportation capability of the gas transmission network is large compared to the installed wind generation, and when power-to-gas is produced at large scale and introduced in major gas arteries (such as at gas terminals), hydrogen input in the grid has a minimal effect on the gas network flow characteristics. More importantly, as a rule, the pressure of the gas supplied to domestic end-use equipment is kept constant, both in the local grid and by means of pressure regulators installed at the entry point of individual houses. However, for specific grid situations in which there is a significant change in pressure upon hydrogen admixture, accounting for the changes in pressure in the fitness for purpose and safety aspects complicates the analysis given below, but is straightforward.

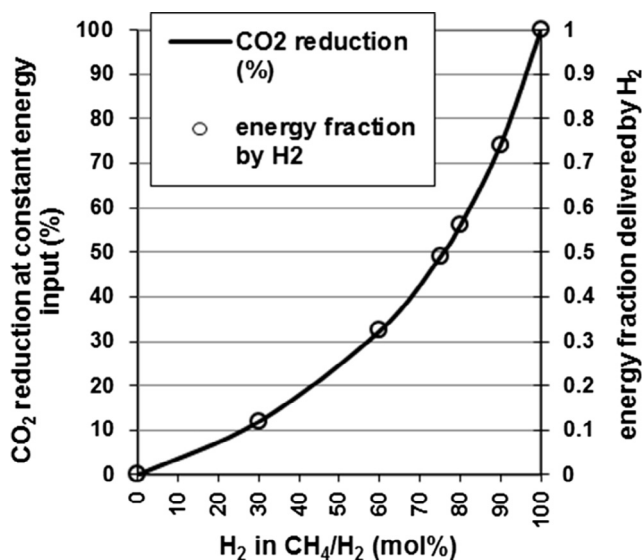


Fig. 1. CO₂ reduction at constant energy input from CH₄/H₂ mixtures, and energy fraction delivered by H₂.

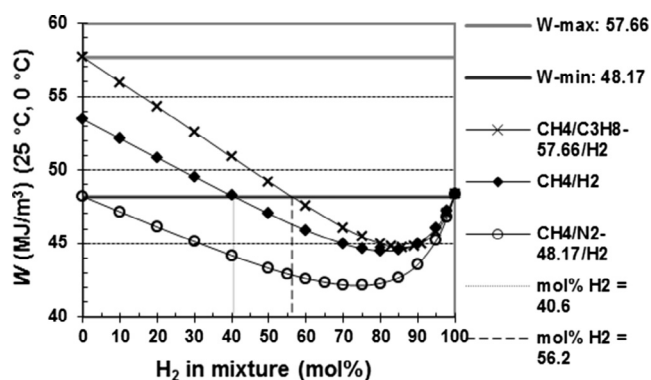


Fig. 2. Wobbe Index as a function of hydrogen admixture to three natural gas compositions that span a fictitious European distribution H-band, $W = 48.17\text{--}57.66$ MJ/m³ (25 °C, 0 °C). (The compositions of the three gases are given in Section 9, below.)

fitness for purpose of different fuel gases. As such, it is often used to characterize the acceptability of fuel gases in common natural gas distribution practice [36,48,49]. Hence, the functional dependence of the changes in Wobbe Index upon hydrogen addition, illustrated for three natural gases in Fig. 2, is determined by the (linear) decrease in heating value of the fuel as discussed above, and the decrease in \sqrt{d} . As seen in Fig. 2, this dependence leads to a minimum in W at $\sim 80\%$ hydrogen in natural gas.

Since the current situation is that natural gases only contain negligible fractions of hydrogen [44,45], the Wobbe Index of natural gas will decrease upon hydrogen addition until relatively high fractions. The question then arises as to what decrease in W , i.e. which decrease in thermal input, is acceptable for the end user. Most countries have a given bandwidth (UK [39], Denmark [50], USA [51], for instance), where only a limited variation is accepted. While the limit of the band is more governed by safety considerations (see below), specifying a minimum Wobbe Index also specifies a minimum thermal input upon which the end user can rely. A lower value adversely affects the fitness for purpose of the fuel.

To see the effects of hydrogen addition to different natural gas compositions, as an example consider a fictitious Wobbe distribution band defined by the so-called European H-band ($W = 48.17\text{--}57.66$ MJ/m³ (25 °C, 0 °C)) [48,49]. While few countries specify such a wide band, it is illustrative to consider it. In a band of distributed natural gases, the end user is ensured that domestic appliances will not have a lower

thermal input than that when fuelled with the lowest Wobbe Index. As a result, when adding hydrogen to natural gas, decreasing the Wobbe Index to a lower value than this limit will provide lower thermal input than what was agreed upon. In Fig. 2, we illustrate this point using three gases that have different Wobbe Indices, similar to the exercise presented in [18]. For this purpose, we consider pure methane in the middle of the fictitious band and two gases at the extremes of the band. Using the lowest Wobbe Index (48.17 MJ/m³) as the limit for adequate thermal input, we see that the highest Wobbe gas can accept the most hydrogen ($\sim 56\%$) before reaching the lower limit, while roughly 41% hydrogen can be admixed to pure methane before reaching the lower Wobbe limit. However, as seen in this figure, the natural gas that is already at the lower limit of the band can admit no hydrogen and still satisfy the requirement for thermal input. Thus, the hydrogen fraction that can be admitted to natural gas while maintaining the performance requirement for thermal input is dependent upon the composition of the natural gas to which hydrogen is added. This point will recur several times below, in the discussion of appliance safety. Clearly, countries having a narrower distribution band can admit less hydrogen.

Enabling hydrogen addition that always satisfies the requirements with varying natural gas composition requires a control method in the grid management system that varies the amount of hydrogen depending on the gas composition.

6. Changes in appliance fuel/air ratio

In almost all domestic appliances the fuel is mixed with a significant amount of air before exiting the burner surface. The degree of pre-mixing, often expressed as the equivalence ratio, is the dominant factor in determining flame behavior [27,40]. The equivalence ratio, Φ , can be written as the ratio of the actual volumetric flow rates of fuel (F) to air (A) divided by its stoichiometric value: $\Phi = (F/A)/(F/A)_{\text{stoich}}$. Here, we take dry air as the oxidizer. Defining the stoichiometric ratio $(F/A)_{\text{stoich}} = 1/L_{dV}$, where L_{dV} is the stoichiometric air requirement, i.e. the volume of air required to oxidize one volume of fuel, we obtain

$$\Phi = (F/A) \times L_{dV}. \quad (2)$$

Possible changes in Φ when changing the fuel composition in domestic appliances have been considered previously [31,41]. In nearly all domestic natural gas appliances, at constant supply pressure the fuel pressure entering the burner is constant independent of the fuel composition, and the momentum of the fuel jet is constant. Under these conditions, in burners using a venturi construction to draw the air into the burner [31,41] the air flow rate will also be constant. For modern, fan-driven, appliances, the volumetric flow of air is only determined by the fan speed, which is also generally independent of the fuel composition.

When the fuel composition is changed at constant supply pressure, the fuel volumetric flow rate F changes according to Eq. (1), while the air flow rate A remains constant. From Eqs. (1) and (2) it follows that changing the fuel composition from fuel 1 to fuel 2 will cause a shift in the equivalence ratio

$$\Delta\Phi = \Phi_2 - \Phi_1 = \Phi_1 \times \left[\left\{ \frac{L_{dV2}}{L_{dV1}} \times \left(\frac{\sqrt{d_1}}{\sqrt{d_2}} \right) \right\} - 1 \right]. \quad (3)$$

The impact of this shift is illustrated in Fig. 3. Here we take pure CH₄ as fuel composition 1 at different initial equivalence ratios and show the effect of increasing H₂ admixture.

The equivalence ratio is seen to decrease monotonically until hydrogen fractions of $\sim 90\%$; that is, the fuel–air mixture becomes leaner. This is a consequence of the progressive decrease in both density and L_{dV} with increasing hydrogen fraction. For instance, fuel-rich combustion at $\Phi_1 = 1.3$ with CH₄ becomes stoichiometric combustion upon admixing about 68% H₂. The size of the shift in equivalence ratio increases with increasing value of Φ_1 , since it is simply proportional to Φ_1 . These shifts have major consequences for the changes in burning velocity, and thus for flashback, as S_L depends strongly on Φ . This will

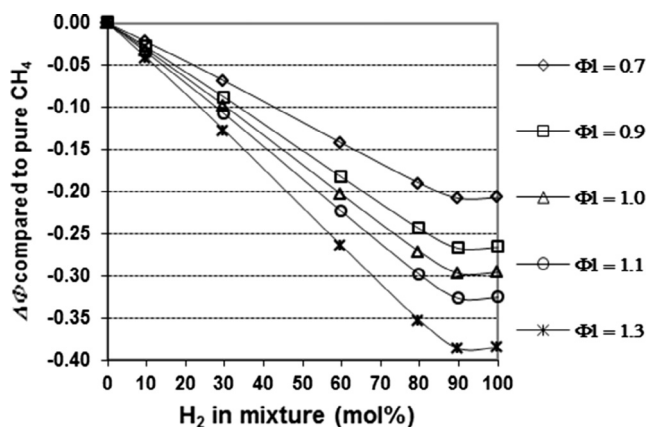


Fig. 3. Shift of equivalence ratio according to Eq. (3), as a function of hydrogen admixture to CH_4 for several initial values of the equivalence ratio ϕ_1 with CH_4 .

be discussed further in Section 7, below.

We note that the effect of the addition of hydrogen on the equivalence ratio is distinct from that resulting from changes in the hydrocarbon ratios of natural gas. There, assuming again that the supply pressure of gas and the air flow to the appliance are constant, $\Phi \propto L_{dV}/\sqrt{d}$. A peculiar property of the lower alkanes comprising natural gas is that the stoichiometric air requirement L_{dV} is proportional to the higher heating value, H_s , to a very good approximation. As a result, the equivalence ratio is de facto proportional to the Wobbe Index. For admixture of H_2 to a natural gas, the relation between heating value and air requirement no longer holds.

7. Analysis of the risk of flashback for domestic appliances using burning velocities

7.1. Effects of shift in equivalence ratio on burning velocity

When the burning velocity in the primary flame front S_L exceeds the velocity v_u of the unburned mixture leaving the burner exit to such an extent that the flame will no longer remain stabilized outside the burner port, the flame will propagate upstream into the burner, ‘flashing back’ into the appliance [27,41,52]. As mentioned above, after flashback, the flame can either continue to burn in the interior of the burner or extinguish. This situation can lead to damage to the burner, appliance shutdown (if a flame safety device is present) or spillage of combustible mixture into the living space, resulting in an explosion hazard. As such, flashback is to be avoided; approval testing and installation rules [48,53] are intended to prevent flashback from occurring during routine appliance use with the range of natural gases to be supplied to the appliances. Actual flashback in individual appliances is a complex function of burner design (equivalence ratio, exit velocities of the unburned fuel–air mixture, burner port geometry, etc.) [41,52], and we note that burners with equivalence ratios close to the stoichiometric value and those with relatively large burner ports [31,41,52] are more likely to flashback than others. Since the focus in this study is on the interchangeability of the fuel, we assess the changes in burning velocity, the parameter characterizing flashback, caused by hydrogen admixture rather than considering individual appliances as installed in the field.

For this purpose, we compute the laminar burning velocity S_L of a number of fuel mixtures; to a good approximation the flow of gas/air mixtures from burner ports in domestic appliances can be considered to be laminar. In light of the discussion above, we explicitly consider the effects of hydrogen admixture on the equivalence ratio. One-dimensional burning velocities are computed using the CHEMKIN software

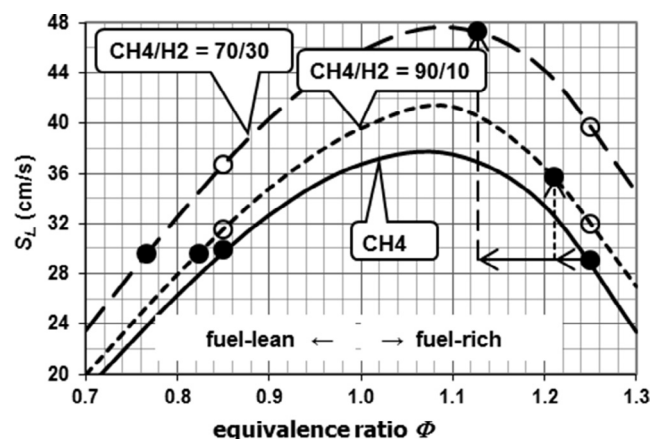


Fig. 4. Behavior of the laminar burning velocity for CH_4 and its mixtures with 10 mol% and 30 mol% of H_2 . Filled symbols indicate burning velocities when considering shifts in equivalence ratio, while the open symbols denote changes in velocity without shifts in equivalence ratio. At the fuel-rich side equivalence ratio shifts and concomitant burning velocities are indicated by solid and dashed arrows, respectively.

and the GRI Mech 3.0 chemical mechanism [54]. For the fuel compositions studied here, the computations faithfully reproduce the trends experimentally observed [55,56].

Fig. 4 shows a portion of the curves of burning velocity as a function of equivalence ratio for 3 mixtures, pure methane and methane with 10% and 30% admixture of hydrogen. The figure shows the expected strong influence of both hydrogen addition and equivalence ratio on the actual burning velocity. While from a physical–chemical perspective it is instructive to consider changes in burning velocity with hydrogen addition at constant equivalence ratio [25], we illustrate the impact of the *shift* in equivalence ratio caused by hydrogen addition below.

Towards this end, we assess the effect of the shift in equivalence ratio on burning velocity for two different equivalence ratios in which we can subdivide the vast majority of domestic appliances: an initial equivalence ratio $\phi_1 = 0.85$, representative of a modern fuel-lean condensing boiler, and $\phi_1 = 1.25$, characteristic of the traditional Bunsen-type appliances, such as cookers, ovens, hot water heaters and non-condensing boilers. We note that different initial equivalence ratios result in the same trends in burning velocity upon hydrogen addition.

For the fuel-lean case ($\phi_1 = 0.85$), Fig. 4 shows that without a shift in equivalence ratio, the burning velocity would increase from ~ 29.5 cm/s for pure methane to roughly 37 cm/s for the 30% mixture (open symbols). However, hydrogen addition shifts the equivalence ratio in a real appliance from 0.85 for pure methane to ~ 0.76 for 30% hydrogen in the fuel mixture (filled symbols), which for these conditions actually results in a slight *decrease* in burning velocity compared to methane. As will be discussed in more detail below, for the fuel-lean case, the increase in burning velocity caused by up to $\sim 50\%$ hydrogen addition will be *largely compensated* by the decrease caused by the concomitant change in appliance equivalence ratio. Thus, hydrogen addition to lean-premixed domestic appliances is expected to have essentially no impact on the flame stability, which has been amply observed in laboratory experiments on this type of domestic appliance [26]. In fact, this example shows that the necessity of empirical observations on lean-premixed appliances is obviated by a relatively simple analysis of burning velocity (flame stability) and Wobbe Index (thermal input).

A substantially different result is obtained for the fuel-rich case shown in Fig. 4. For a $\text{CH}_4/\text{H}_2 = 70/30$ mixture the increase in burning velocity at constant equivalence ratio would be about 10 cm/s (35%) (open symbols), while the shift in equivalence ratio (horizontal arrows) results in an increase of more than 18 cm/s (61%) (vertical arrows,

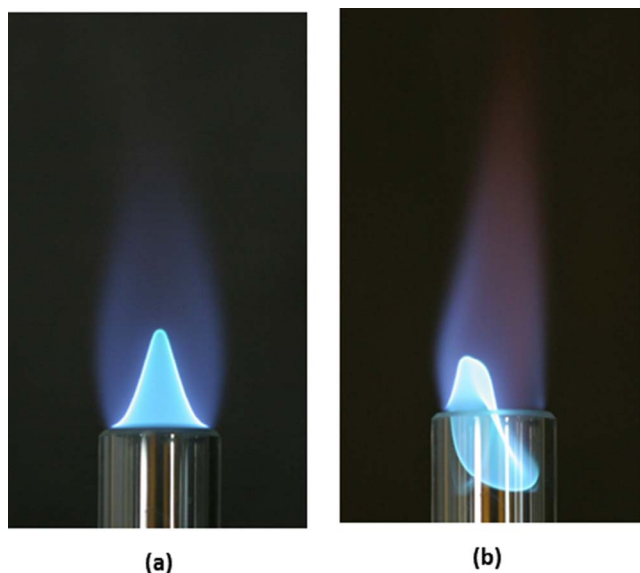


Fig. 5. Bunsen tube (1 cm inner diameter) experiments to substantiate the results of the burning velocity calculations. (a) Stable free flame situation. (b) Flame just flashing back upon lowering the flow velocity of fresh gas/air mixture from the stable free flame situation.

filled symbols). Therefore, under fuel-rich conditions the shift in equivalence ratio *amplifies* the effect of hydrogen on burning velocity.

7.2. Use of burning velocities to scale the tendency for flashback: comparison with experiments

While the use of one-dimensional burning velocities, including their dependence on equivalence ratio, to compare the effects of fuel composition is straightforward and, as seen above, capable of describing the trends observed in domestic appliances, more quantitative analysis requires additional consideration. In [27], the physical analysis of flashback showed that in general flashback occurs when the burning velocity exceeds the exit velocity everywhere in the flow. This approach allows the analysis of essentially all burner types, but not without significant numerical or experimental effort for the two- or three-dimensional flow fields in many domestic appliances. As stated above, we avoid the consideration of the vagaries of individual appliances and assess the degree to which the use of burning velocities is warranted to predict the limits of flashback when changing fuel composition in a multi-dimensional geometry. For this purpose, we performed experiments using Bunsen tubes, where for a given fuel–air mixture we create conditions in which the burning velocity is greater than the exit velocity, causing flashback. These burners have the advantage that the equivalence ratio and exit velocity can be carefully varied and reproduced; this is not possible in most domestic appliances. Furthermore, it should be recalled that many standards for gas interchangeability have been determined using these tubes [32].

Here we use a quartz tube of 1 m length and 1 cm inner diameter. For the experiments, well-developed laminar parabolic flow is obtained for all flow velocities reported here. Gas and air flows are metered by calibrated massflow controllers (Bronkhorst); the controllers were coupled by computer to permit varying the total flow of fuel–air mixture through the burner while maintaining a preset value of the equivalence ratio. Mixing of fuel and air is accomplished in a static mixing facility before entering the quartz tube. The equivalence ratio was verified by sampling a small fraction of the fuel–air mixture after each experiment and analyzing it in a methane meter (Maihak Unor). After igniting a stable flame (exit velocity > burning velocity), as illustrated in Fig. 5a, the exit velocity was reduced by reducing the total flow until flashback occurred. Flashback was taken when the flame just

entered the burner tube (Fig. 5b); this point was very reproducible and differed with complete flashback (where the flame propagates upstream within the tube) only marginally [27,57,58]. The measurements were reproduced by extinguishing the flame, allowing the tube to return to room temperature, and resetting the initial exit velocity. For each gas composition, the experiment was repeated 10 times. The equivalence ratios were reproducible to within $\pm 1\%$, while the flows of fuel–air mixture at flashback are reproducible within $\pm 0.7\%$. The hydrocarbon/nitrogen/hydrogen fuel mixtures used in the experiments (see below) were obtained commercially as mixtures, with compositions determined by gas chromatography to better than $\pm 1\%$.

While the well-defined conditions of the experiment allow the comparison of the burning velocity of the mixture with the local flow velocities at flashback, our interest here is in assessing whether the burning velocity can be used to compare the flashback propensity of different fuel compositions. For these purposes, we characterize the velocity at flashback as the average flow velocity of the unburned mixture at that point $v_{u,f}$, obtained as the volumetric flow rate divided by the cross-sectional area of the tube.

With an eye towards understanding the impact of the natural gas composition to which hydrogen is added, we consider admixture of hydrogen to three different ‘natural gases’, whose range of Wobbe Index roughly spans the variations in the EU. A relatively low-Wobbe gas $CH_4/N_2 = 92.5/7.5$ ($W = 48.12 \text{ MJ/m}^3$),³ an intermediate natural gas (pure methane, $W = 53.45 \text{ MJ/m}^3$) and a gas with a relatively high Wobbe Index $CH_4/C_2H_6 = 80/20$ ($W = 56.78 \text{ MJ/m}^3$). As described above, the risk of flashback in domestic appliances upon hydrogen addition is greatest for those having fuel-rich ($\Phi > 1$) Bunsen-type flames. For the comparison considered here, we take the equivalence ratio for the middle gas, pure methane, without hydrogen addition at $\Phi = 1.3$; for all other mixtures, the equivalence ratio was shifted in accordance with its composition, following the method described in Section 6, above. Both the experiments and the computations allowed for this shift in equivalence ratio. The results of this exercise are presented in Fig. 6. The upper curves (solid symbols) show the $v_{u,f}$ for the different fuel mixtures, while the lower curves are the computed burning velocities (S_L).

Since $v_{u,f}$ is an average quantity expected to be proportional to the burning velocity, but not identical to it, the computed burning velocities and the measured average exit velocities have different numerical values, as observed by the shift in the data along the velocity axis in Fig. 6. We observe that despite this shift the experimental and computational data show the same qualitative ranking for the three natural gases along the velocity axis for the range of hydrogen admixture studied (up to 40% H_2 in the mixture).

Using the notion of a reference fuel, we can quantitatively rank the flashback propensity of the gases and also compare the experimentally determined $v_{u,f}$ and the computed S_L for the different natural gases, to ascertain whether the same ranking is obtained in terms of hydrogen admixture. As a reference point in these comparisons, we choose a gas composition that is assumed to cause flashback in the range of gas compositions being studied. If the measured exit velocity of a fuel mixture at flashback is higher than that of the reference gas, this means the burning velocity is higher, and the fuel mixture will cause flashback more readily than the reference gas. The horizontal lines are the measured and computed values for the reference fuel; here taken as $CH_4/H_2 = 77/23$ ($W = 50.44 \text{ MJ/m}^3$).

If we examine the point at which the propensity for flashback is the same as for the reference fuel, i.e., where the curves intersect the lines for the reference fuel, we see that this point is very well predicted by

³ Realizing that the Wobbe Index of this base gas is already at the lower limit, which would prevent any hydrogen addition at all, for the relatively low-Wobbe gas, upon hydrogen admixture the mixture Wobbe Index was kept nearly constant by simultaneously increasing the CH_4/N_2 ratio.

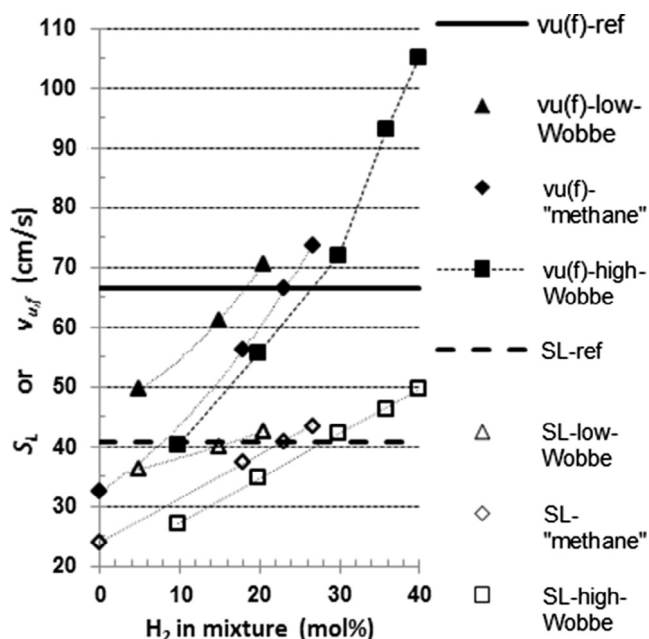


Fig. 6. Unburned mixture flow velocities at flashback, characterized by the experimentally determined average $v_{u,f}$ and calculated burning velocities S_L , for three natural gases upon H_2 admixture. The horizontals are the measured and computed values for the reference fuel $CH_4/H_2 = 77/23$.

the burning velocities, differing by at most 1.8% in H_2 fraction ($< 7\%$ relative difference), which is within the experimental uncertainty. Since the experimentally determined hydrogen fraction and that predicted by the burning velocity calculations give the same results within the uncertainty of the experiments, we conclude that we can use the burning velocities to predict flashback in comparison with a reference gas. As a consequence, for the rest of the discussion we use the burning velocities to predict flashback propensities for natural gases with increasing amounts of hydrogen admixture.

The salient observation from Fig. 6 is that for the three different natural gas compositions the hydrogen fraction at which the flashback propensity is equivalent to that of the reference gas is different, with these amounts of hydrogen admixture increasing with the Wobbe Index of the NG to which it is added. Thus, compared to methane, for the low Wobbe gas, 5% less hydrogen can be in the gas before it flashes back at the same exit velocity as the reference gas, while for the high Wobbe gas roughly 3% more hydrogen can be added. As will be discussed in detail below, this variation is caused by the shift in equivalence ratio arising from the composition of the natural gas itself. *That the amount of hydrogen admixture resulting in a given reference level of flashback depends on the composition of the natural gas to which hydrogen is added is an essential point that arises in all interchangeability assessments* (Sections 9 and 10, below). We also observe that both the experimentally and computationally determined fraction of hydrogen in methane at flashback is within 1% of the 23% used for the reference gas, giving further confidence in the internal consistency of the method.

8. Assessing the risk of flashback for appliance populations

The exercise performed in Section 7.2 illustrates the application of burning velocities to assess flashback limits from hydrogen admixture for one class of burner. To perform interchangeability assessments on NG/ H_2 mixtures for a population of installed appliances using this method, we must consider the variation in equivalence ratio in such a population. While the exercise can be performed for all possible equivalence ratios, we can make a few simplifying assumptions without loss of generality. First, the S_L -dependence on ϕ , as discussed around Fig. 4, above, shows that fueling the fraction of the appliance

population that is lean-premixed ($\phi < 1$, most modern condensing boilers) with hydrogen-containing mixtures is expected to have essentially no impact on the flame stability. As a result, we can focus the study more on the impact on the fuel-rich part of the population, e.g., hot water heaters, conventional domestic boilers and furnaces, cookers etc., which is more sensitive to flashback. The actual values of the equivalence ratio used for appliance adjustment (ϕ_{adj}) and ϕ -shifts determine the outcome of the exercise.

The next assumption regards the choice of the reference gas, de facto choosing the limiting requirements for flashback. To the authors' knowledge there have been no systematic studies investigating the actual changes in the response of domestic appliances to varying gas quality during the lifetime of the appliances [53,59]. Just because an appliance could accommodate a given gas without flashback when it was new, does not mean it can do so when having been in practical use for many years, particularly when considering the large variation in the state of maintenance in most populations. Rather than assume the robustness, or lack thereof, of the appliance population directly, we imply the sensitivity of appliances to variations in gas composition based on distribution practice: that the appliance population, with all conditions of age and maintenance, can safely accept the range of gases currently being distributed, as has been done previously in a practical interchangeability situation [36]. Therefore, an interchangeability assessment of flashback requires determining the limiting burning velocity that characterizes flashback, $S_L(\phi)_{w.c.}$, indicating the *worst case for the current practice of natural gas distribution*, in other words the highest risk of flashback for the installed population. Referring to Fig. 6, if appliances are considered to operate properly (not flash back) when fueled with the natural gas having the highest propensity for flashback (highest burning velocity), then maintaining the burning velocities of H_2/NG mixtures below this level guarantees that the risk of flashback will not increase upon hydrogen admixture.

When considering the impact of natural gas composition on flashback, we recall that the largest changes in burning velocity are caused by possible changes in equivalence ratio. As discussed in Section 6, above, for natural gases the primary equivalence ratio is proportional to the Wobbe Index. Referring to Fig. 4, above, for fuel-rich appliances, gases supplied at the lower end of the Wobbe range will shift the burning velocity farthest towards the stoichiometric value, causing the largest increase. Thus, assuming all appliances function properly with the lowest distribution gas, this gas will cause the highest burning velocity in the population of appliances and can serve as a reference gas for flashback. In our fictitious natural gas distribution band (Section 5), this gas has a Wobbe Index 48.17 MJ/m^3 .

Having defined the reference natural gas for flashback, given the impact of equivalence ratio and the shift caused by hydrogen addition, the remaining assumption to be made for the purposes of illustrating the response of appliances to the risk of flashback is the choice of equivalence ratio representative of the installed appliance population. Again, rather than computing the changes in burning velocity to quantify the change in risk of flashback for all (fuel-rich) equivalence ratios, here we use a worst-case appliance adjustment (ϕ_{adj}), defined for a specific 'adjustment gas'. We note that the primary equivalence ratio of fuel-rich appliances generally cannot be adjusted in the field [29] and, for the fictitious distribution band considered here, these appliances can be assumed to be adjusted by the manufacturer with methane, the 'adjustment gas' for the European H-band [48]. Clearly, as can be seen in Fig. 4, above, and considering the natural gas chosen as a reference gas for flashback, an appliance that is adjusted at an equivalence ratio closer to the stoichiometric point will flash back at a lower hydrogen fraction than at a higher equivalence ratio. In general, appliances are adjusted to suit wide variations in conditions [41] and, regarding flame stability, the equivalence ratio is chosen to avoid both flashback and flame lift. For fuel-rich appliances, the risk of both phenomena increases when approaching the stoichiometric value [41]. For very high equivalence ratios, $\phi > 1.6$, the changes in burning velocity

are such that the normal variation in natural gas composition will never result in flashback. As such, this range of equivalence ratio is considered relatively insensitive to hydrogen addition, and appliances with lower equivalence ratios will be earlier at risk. For the purposes of the exercise performed here, we assume as a worst case that appliances are adjusted with pure methane at $\Phi_{adj} = 1.25$, which is just outside the region of equivalence ratio for which flashback can readily be observed [41].

Having defined a reference gas for flashback, an adjustment gas and an adjustment equivalence ratio, we can now quantify the impact of hydrogen admixture to natural gas on appliances.

9. Admixing H₂ in natural gas distribution

Using our fictitious distribution band as an example, hydrogen admixture will be considered for three model natural gases described above (Fig. 2): a natural gas at the upper limit of the band $W = W_{max} = 57.66 \text{ MJ/m}^3$, having the composition⁴ CH₄ = 87 mol%, C₃H₈ = 13 mol%, pure methane ($W = 53.45 \text{ MJ/m}^3$), and a natural gas at the lower limit of the band $W = W_{min} = 48.17 \text{ MJ/m}^3$ having composition CH₄ = 92.6 mol%, N₂ = 7.4 mol%. In the discussion below, we denote these three gases as W_{max} , CH₄ and W_{min} , respectively.

The flashback behavior upon hydrogen admixture of fuel-rich vs. fuel-lean appliances will be contrasted in detail. In some modern lean-premixed central heating boilers the gas/air ratio is controlled to remain constant upon changing gas composition. When relevant, considering, for example, the actual numbers of appliances in use, this situation can be easily assessed.

9.1. Fuel-rich appliances

The computed burning velocities as a function of equivalence ratio, $S_L(\Phi)$, for the three model natural gases, and the effects of H₂ admixture are shown in Fig. 7.

The starting point is the appliance adjustment with the adjustment gas CH₄ at equivalence ratio $\Phi_{adj} = 1.25$, as explained in Section 8, above (the filled black diamond in Fig. 7). The effect of the shift in equivalence ratio when substituting the different natural gases for methane within the band is shown in the figure. Changing from methane to the distribution gas with the lowest Wobbe Index (“ W_{min} ” in the figure) shifts the equivalence ratio from 1.25 for methane to 1.13 (the solid horizontal black arrow) and shifts the burning velocity from 29.4 cm/s to 36.4 cm/s (open blue circle and vertical dashed black arrow in the Figure). Thus, the ‘leaner’ gas (lower Wobbe Index) has a burning velocity that is roughly 25% higher than CH₄ at the same appliance conditions, caused by the Φ shift, while at constant equivalence ratio the leaner gas has burning velocities lower than CH₄. Since the leaner gas was chosen to be the reference gas for flashback, the burning velocity obtained by substituting this gas for methane at this equivalence ratio is the ‘worst case’ burning velocity, 36.4 cm/s. As discussed above, this gas forms the limit for risk, and this burning velocity is indicated by the red solid horizontal line in Fig. 7. Switching to the gas with the highest Wobbe Index, W_{max} , yields a positive Φ shift, as indicated by the solid horizontal black arrow in Fig. 7. The vertical dashed black arrow points to the lowest flashback propensity for this gas (brown x in the Figure) with $S_L = 21.9 \text{ cm/s}$.

Considering hydrogen addition, the shifts in equivalence ratio and increasing values of burning velocity for the different natural gases result in trajectories with increasing amounts of hydrogen, as indicated by the green dotted trajectories in the Figure. As will be shown

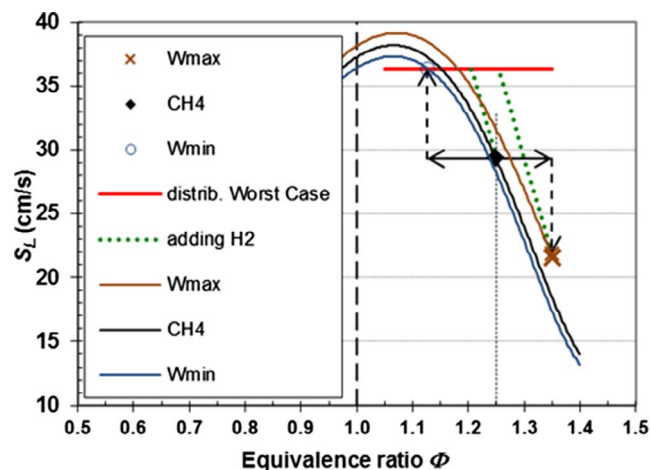


Fig. 7. Burning velocity behavior as a function of equivalence ratio for the three natural gases considered, in the fuel-rich domain, indicated by the curves. The appliance adjustment ($\Phi_{adj} = 1.25$) is given by the solid diamond. The equivalence ratio shifts and resulting burning velocities for the other two natural gases are indicated by solid and dashed arrows, respectively. The green dotted trajectories, labeled “adding H₂”, show the resultant changes in equivalence ratio and burning velocity upon adding hydrogen to the point at which the burning velocities are equal to that of the reference gas at $W_{min} = 48.17 \text{ MJ/m}^3$ (red line). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

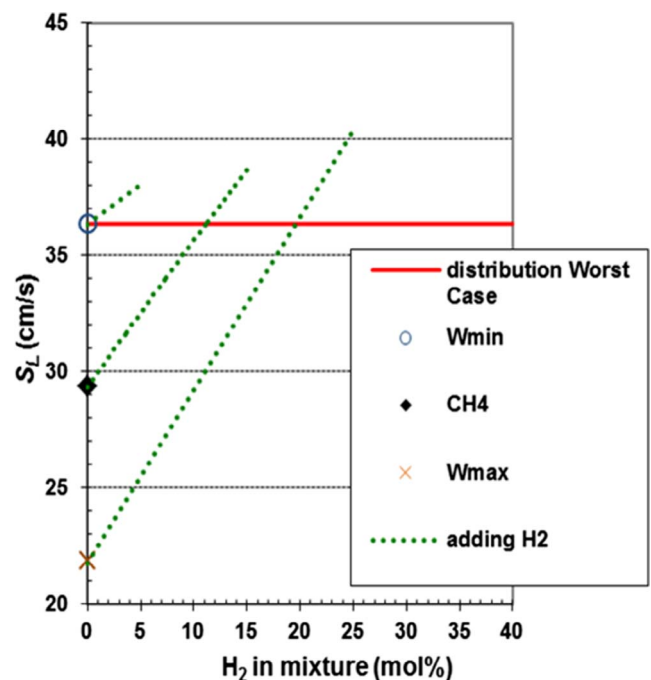


Fig. 8. Burning velocity for the three natural gases, in the fuel-rich domain, upon admixing hydrogen.

explicitly in Fig. 8, below, since the interchangeability criterion for flashback says that the maximum burning velocity may not be higher than that of the reference gas for flashback, in the fuel-rich domain W_{min} does not allow any H₂ admixture, while CH₄ and W_{max} both allow substantial H₂ fractions before reaching the maximum allowable burning velocity for the reference gas (the red line).

The change in computed burning velocity with hydrogen admixture for each of the three gases is shown in Fig. 8, and compared with the limit burning velocity for the reference gas. As mentioned above, the gas with the lowest Wobbe Index, taken as the limit, admits no hydrogen admixture. For the other gases, 11.2 mol% can be admixed into pure methane, and 19.7 mol% can be admixed into W_{max} before the

⁴ The compositions have been rounded off for convenience. We also note that, given the dominant effects of hydrogen admixture on the burning velocity, using different natural gas compositions for the different Wobbe Indices has a negligible impact on the results.

limiting burning velocity is reached: thus, *these are the fractions of hydrogen that can be admixed to these gases without an increased risk of flashback.*

We note here that, recalling the discussion in Section 6, adding hydrogen to natural gas will also change the exit velocity of the fuel air mixture, because the density of the H₂/NG mixture will decrease. Since flashback is a competition between burning velocity and exit velocity [24], the expectation is that the increase in exit velocity will mitigate the increase in burning velocity to some extent. In this case, using $S_L/(F + A)$ as an interchangeability parameter is more correct. Performing the same exercise as above using this parameter rather than the burning velocity alone changes the results by less than 1% in maximum hydrogen fraction. As such, we neglect it in further considerations of the fuel-rich domain.

It is important to emphasize here the precise meaning of these interchangeability results. The intention is to determine how much hydrogen can be admixed in natural gas in the existing population of domestic appliances with no added risk of flashback, without the necessity of appliance readjustment and explicitly without considering the performance of individual appliances. An interchangeability result of a maximum of say 10% hydrogen addition to a given natural gas does not mean that at higher fractions installed appliances will start to flash back (since we assume no knowledge about the individual appliances); it only means that the *risk* of flashback increases above that for the normal practice of natural gas distribution; *the performance of the appliance population cannot be guaranteed* at fractions of more than 10% in this example.

The assessment thus far computes the maximum hydrogen admixture starting from a specific natural gas composition. Since it may not always be convenient in distribution practice to adjust the flow of hydrogen based on the composition of natural gas flowing past the injection point, we can also ask the question as to which part of the natural gas distribution band would allow a certain constant hydrogen fraction, again with the constraint of using the distribution Wobbe limit as the worst case for flashback. Here we consider the range of Wobbe Index for which 10 mol% hydrogen does not increase the risk of flashback. Since we know from Fig. 8 that pure methane can admit more than 10 mol% hydrogen, only the region 48.17–53.45 MJ/m³ is considered, and we progressively add nitrogen to methane to lower the Wobbe Index of the natural gas before adding the 10% hydrogen. The results are shown in Fig. 9. For all natural gases with $W \geq 52.9$ MJ/m³, the burning velocities of the corresponding mixture with 10 mol% H₂

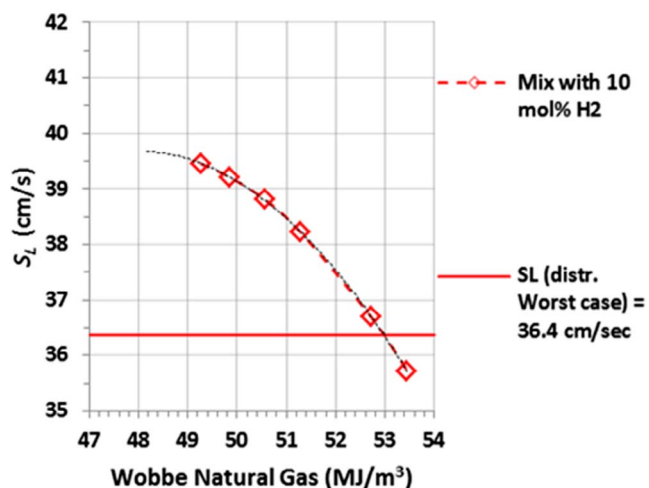


Fig. 9. Burning velocity of CH₄/N₂/H₂ mixtures containing 10 mol% of H₂, as a function of the Wobbe Index of the corresponding natural gases without hydrogen. Open diamonds are the burning velocities of individual fuel mixtures. The solid red line denotes the burning velocities of W_{min}. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

(of course, allowing for the shift in equivalence ratio) are equal to, or below that of, the reference gas for flashback. Thus, 10 mol% hydrogen admixture in these gases causes no increase in risk for the appliance population, but for $W < 52.9$ MJ/m³, 10 mol% represents an increased risk. For natural gases with $W \leq 49.26$ MJ/m³, the Wobbe Index W of the corresponding mixture with 10 mol% H₂ decreases below the lower Wobbe limit for the distribution band, $W_{min} = 48.17$ MJ/m³. Simulating the upper part of the Wobbe band 53.45–57.66 MJ/m³ using CH₄/C₃H₈ mixtures confirmed the expected result that 10 mol% H₂ does not increase the risk for natural gases in this range, and the Wobbe Index remains within the distribution range. For our fictitious distribution band, the overall result for the compositions considered is that admixture of 10 mol% H₂ is acceptable in the upper part of the band, in the range $52.9 \text{ MJ/m}^3 \leq W_{NG} \leq 57.66 \text{ MJ/m}^3$, from the viewpoints of both the Wobbe Index and the risk of flashback in domestic appliances.

9.2. The impact of hydrogen admixture for fuel-lean appliances

As indicated above, much of the discussion regarding the impact of NG/H₂ mixtures on domestic appliances makes reference to experiments on modern fuel-lean appliances. In Section 7.1 it was shown that for the fuel-lean case, the increase in burning velocity upon hydrogen addition up to 40–50 mol% H₂ will be compensated by the effect of the shift in equivalence ratio. Although fuel-lean appliances are not expected to constitute a critical part of the overall appliance population regarding flashback, it is still illustrative to quantify the risk of flashback upon hydrogen admixture in the fuel-lean domain, to contrast results from the fuel-rich domain.

The curves of burning velocity versus equivalence ratio in the fuel-lean domain are shown in Fig. 10. In this case a general appliance adjustment is considered with CH₄ at $\phi_{adj} = 0.85$ (filled black diamond in Fig. 10). For a distribution range of natural gases supplied to a fuel-lean appliance, the maximum burning velocity, and thus the maximum risk of flashback, occurs using the gas with the *highest* Wobbe Index, W_{max} , which is the reference gas for flashback for these appliances. The shift in equivalence ratio from the adjustment point (filled diamond) caused by the increase in Wobbe Index (solid black arrow in Fig. 10)

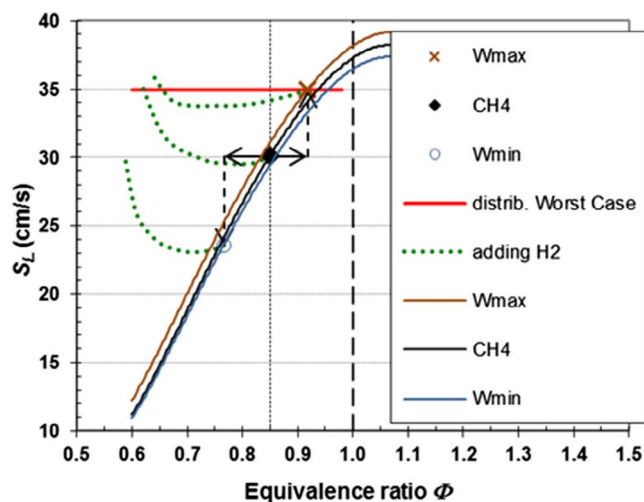


Fig. 10. Burning velocity behavior as a function of equivalence ratio for the three natural gases considered, in the fuel-lean domain, indicated by the solid curves. The solid black diamond indicates the appliance adjustment at $\phi_{adj} = 0.85$ with CH₄. The equivalence ratio shifts and resulting burning velocities for the other two natural gases are indicated by solid and dashed arrows, respectively. The green dotted trajectories, labeled “adding H₂”, show the resultant changes in equivalence ratio and burning velocity upon adding hydrogen to the point at which the burning velocities are equal to that of the reference gas W_{max} . (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

increases the burning velocity (dashed arrow) to the ‘worst case’ value, $S_L = 35.0$ cm/s (brown cross in Fig. 10). Here, the *richer* gas has a burning velocity that is roughly 16% higher than CH_4 with the same appliance conditions. Switching to the leanest gas, W_{\min} , yields the lowest burning velocity in the distribution band, $S_L = 23.6$ cm/s.

Upon hydrogen addition, the equivalence ratio shifts to lower values, as discussed in detail above (Fig. 3). The progressive changes in burning velocity with hydrogen admixture are shown as a function of equivalence ratio in Fig. 10 (dotted green curves), and as a function of hydrogen fraction in Fig. 11. The contrast with the fuel-rich case is apparent. In Figs. 7 and 8 the dotted green trajectories are monotonically (linearly) increasing, while the fuel-lean case of Figs. 10 and 11 shows substantially different behavior. These curves reflect the competition between the increase in burning velocity caused by hydrogen addition and the decrease in burning velocity caused by the progressive decrease in equivalence ratio. As a result, the fuel-lean case allows a substantial hydrogen admixture to the ‘worst case’ natural gas for flashback, W_{\max} , up to roughly 70%. As shown in Fig. 11, all natural gases within our fictitious distribution band allow large amounts of H_2 admixture in the fuel-lean domain, when using the 35.0 cm/s worst-case burning velocity. Moreover, all these natural gases allow H_2 admixture up to > 40 mol% before the burning velocity will have increased above its original value for the natural gas without hydrogen admixture, further underlining the conclusion that this part of the appliance population is not critical with respect to flashback.

From Fig. 11, the hydrogen fractions that can be admixed without an increase in risk are > 80 mol%, 79.5 mol% and 74.6 mol%, respectively, with W_{\min} , CH_4 and W_{\max} , respectively. We note that above ~70% hydrogen the combustion properties of the mixture begin more to resemble those of pure hydrogen with the admixture of (small quantities of) natural gas than of natural gas with a hydrogen additive. This is manifest in the sharp increase in burning velocity in Figs. 10 and 11 at higher hydrogen fractions.

In contrast with the fuel-rich case, for fuel-lean appliances using $S_L/(F + A)$ as a basis for interchangeability rather than S_L (Section 9.1) changes the maximum hydrogen fraction by several mol%. However, given their high level of tolerance towards flashback for hydrogen in natural gas (> 70 mol%) this difference is not discussed further here.

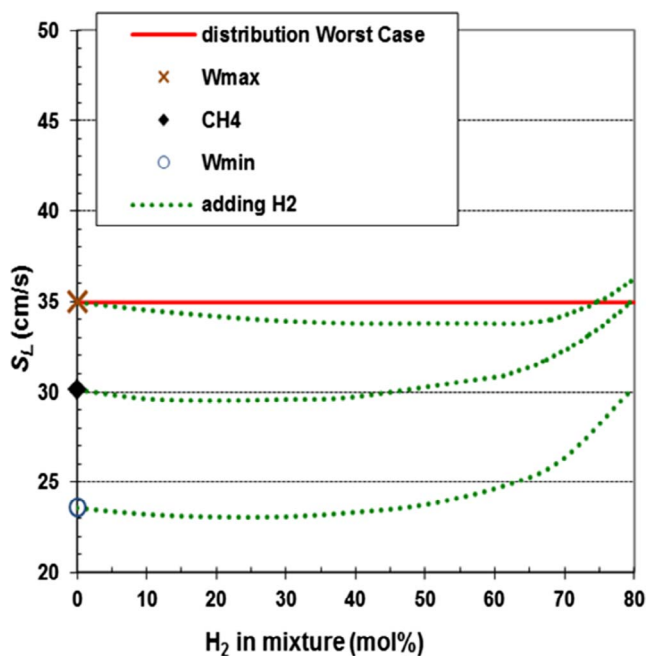


Fig. 11. Burning velocity for the three natural gases, in the fuel-lean domain, upon admixing hydrogen.

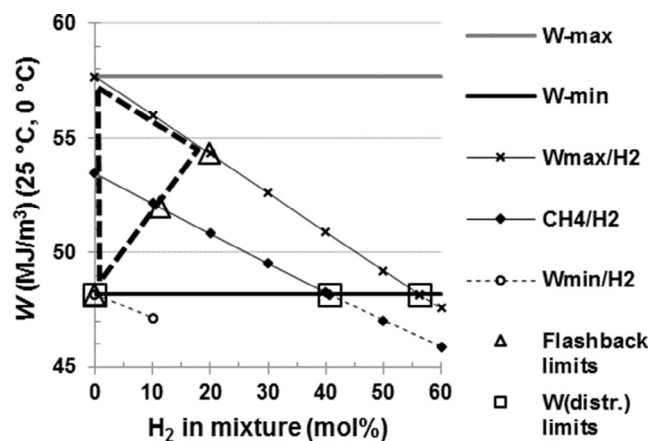


Fig. 12. Working Area for H_2 admixture. The three squares indicated with “W(distr.) limits” represent the limits of admixture as set by the distribution minimum $W_{\min} = 48.17$ MJ/m³. The three triangles indicated with “Flashback limits” represent the limits of admixture as set by the reference gas for flashback for the fuel-rich domain. The overall acceptable range of natural gases and their concomitant hydrogen admixtures is covered by the area within the bold dashed triangle.

10. Working area for H_2 admixture for domestic appliances

Combining the results from Figs. 2, 8 and 11, a simple picture showing the possibilities for H_2 admixture within the limitations of fitness for purpose and safety, i.e., Wobbe Index and flashback, respectively, sketches the situation at a glance. In principle, all appliances (fuel-rich, fuel-lean) are connected to a single conceptual grid. As discussed above, in the fictitious distribution situation considered flashback is only a limitation for fuel-rich appliances. For the three model natural gases, W_{\min} , CH_4 and W_{\max} , and appliance adjustment at $\Phi_{\text{adj}} = 1.25$ using methane, the maximum flashback-limited hydrogen fractions are 0, 11.2, and 19.7 mol%, respectively, while the Wobbe distribution limited values amount to 0, 40.6, and 56.2 mol%, respectively, for these gases. Thus, the fuel-rich portion of the appliance population is limited by flashback, while the fuel-lean appliance population is limited by the constraints on Wobbe Index.⁵ These results are summarized in a ‘Working Area’ diagram, Fig. 12, for the fictitious range of natural gas composition studied. The overall area of mixtures satisfying both Wobbe and flashback constraints is roughly indicated by the bold dashed triangle.

Following the procedure delineated above, more accurate boundaries of the working area can be obtained by assessing hydrogen admixture to the range of natural gas compositions representing the actual range of Wobbe Indices and compositions for a given distribution situation. A local situation having narrower limits on Wobbe Index will give different results. The Netherlands, for example, has a relatively narrow distribution band (a width of 1 MJ/m³, rather than the 9 MJ/m³ considered here). The analysis for this situation shows a flashback-limited maximum hydrogen fraction of only a few mol% [60].

We emphasize here that, as shown in Fig. 12, the analysis presented above demonstrates that the maximum hydrogen fraction depends on the composition of the natural gas to which it is added.

11. Summary and conclusions

In this paper, limits to hydrogen addition formed by considerations of fitness for purpose and safety are derived. The fitness-for-purpose limits are governed by changes in thermal input caused by changes in Wobbe Index upon hydrogen addition, while the changes in risk of

⁵ We recall that we assumed that fuel-lean appliances do not have active fuel–air ratio controls. Appliances with controls are more susceptible to flashback than considered here (compare Fig. 4). Including them in the analysis is straightforward.

flashback are used to quantify the safety aspects. Toward this end, a computational interchangeability analysis has been developed to quantify the risk of flashback in populations of domestic appliances installed in the field. The method used combines the simple description of appliance response to changes in gas composition, which has been amply experimentally verified, with computations of burning velocity. Comparison of flashback measured using natural gas/hydrogen flames on a Bunsen tube with the results of the computational analysis using burning velocity calculations, showed excellent agreement for the flashback-limited maximum hydrogen fractions obtained. The contrasting flashback propensity upon hydrogen admixture of fuel-rich vs. fuel-lean appliances is considered. The analysis shows fuel-rich pre-mixed appliances to be critical for flashback, while fuel-lean appliances are demonstrated to be relatively insensitive for flashback as a matter of principle. Consequently, testing modern fuel-lean appliances not having an active fuel–air ratio control system has no added value with respect to determining the limiting factors for the introduction of H₂ into the natural gas grid.

The results demonstrate that the maximum admixture hydrogen to natural gas depends on the composition of the natural gas to which hydrogen is being added, both in terms of respecting the constraints on the Wobbe Index for a given distribution range and the impact on flashback. This observation will add another dimension of complexity to grid-management schemes for hydrogen addition. The analysis was performed using a fictitious distribution range and can (and should) be performed for the local variations in natural gas composition, regionally or nationally, to assess the actual impact on fitness for purpose and the safety risk from hydrogen admixture.

The insights presented are useful in defining efficient strategic roadmaps, and assessment and testing programs, aimed at introducing hydrogen into the natural gas infrastructure without increasing the risk for the domestic end user [11], as today the maximum hydrogen fraction is still an open issue [19–21]. The physical–chemical basis of the analysis, and the assumptions regarding the choices of reference gases, yield results that are not sensitive to the vagaries of the performance of individual appliances or their state of repair or maintenance. Thus, the risk of flashback in populations of domestic appliances installed in practice can be quantified without the necessity of large-scale appliance testing. This marks an essential reduction of the knowledge gap with respect to appliances' hydrogen tolerance that can be used for assessment of gas grid management when considering the possibility of hydrogen injection. As such, the method can contribute significantly to the assessment of the ultimate role of power-to-gas [6,7].

Applying the approach used here enables a fundamentally justified incorporation of the risk of flashback upon hydrogen admixture in standards on gas quality, such as the German G 260 [61] and G 262 [62] and the GS(M)R [39] in the United Kingdom.

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