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Abstract: The blending of hydrogen gas into natural gas pipelines is an effective way of achieving the goal of carbon neutrality. Due to the large differences in the calorific values of natural gas from different sources, the calorific value of natural gas after mixing with hydrogen may not meet the quality requirements of natural gas, and the quality of natural gas entering long-distance natural gas and urban gas pipelines also has different requirements. Therefore, it is necessary to study the effect of multiple gas sources and different pipe network types on the differences in the calorific values of natural gas following hydrogen admixing. In this regard, this study aimed to determine the quality requirements and proportions of hydrogen-mixed gas in natural gas pipelines at home and abroad, and systematically determined the quality requirements for natural gas entering both long-distance natural gas and urban gas pipelines in combination with national standards. Taking the real calorific values of the gas supply cycle of seven atmospheric sources as an example, the calorific and Wobbe Index values for different hydrogen admixture ratios in a one-year cycle were calculated. The results showed that under the requirement of natural gas interchangeability, there were great differences in the proportions of natural gas mixed with hydrogen from different gas sources. When determining the proportion of hydrogen mixed with natural gas, both the factors of different gas sources and the factors of the gas supply cycle should be considered.

Keywords: natural gas; hydrogen injection; pipelines transport; interchangeability

1. Introduction

Injecting hydrogen into natural gas pipelines not only solves the transmission problem of hydrogen but also deals with the large-scale application issue in the use of hydrogen as an energy source, which also becomes an efficient way of achieving the goal of carbon neutrality. Currently, many western countries are working on the mixed transportation of hydrogen–natural gas mixtures without adjusting the available pipeline facilities [1]. Natural gas quality is the most basic requirement for determining the maximum fraction of hydrogen that can be added to pipeline networks. Because of the high ratios of hydrogen injected, the gas quality needs to satisfy the national standards of various countries. Hydrogen compressed natural gas (HCNG) is unreliable in both long-distance transportation and urban gas pipeline networks. Therefore, it is valuable to consider factors such as pipe material safety and equipment bearing capacity only after the critical proportion of hydrogen for use in HCNG has been determined.

According to Internation Energy Agency (IEA) data, by the end of 2019, there were 37 HCNG demonstration projects in operation around the world [2–5], including the "NaturalHy" project in the European Union (EU), the "Hydeploy" and "H21 Leeds CityGate" projects in the United Kingdom (UK), the "DVG" project in Germany, and the "HIGG"



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program in the US National Fuel Cell Research Center. As shown in Figure 1, the injected hydrogen ratios these projects studied ranged from 5% to 20%.

Figure 1. Mole percentage of hydrogen injected into natural gas pipelines, as studied by different countries' demonstration projects.

Natural gas quality is reflected mainly in calorific value and gas composition. Although the calorific value per unit mass of hydrogen is very high, the calorific value per unit volume is relatively low, which is 11.89 MJm^{-3} under standard conditions (~1/3 of natural gas). Since increasing hydrogen ratios reduce the calorific value of HCNG, the hydrogen fraction directly impacts the quality of transported mixed gases. Moreover, the composition and calorific value of natural gas also vary across different regions and in different supply cycles. For example, Japanese natural gas is imported mainly by the liquefied natural gas (LNG) method, and has a more mid-range calorific value. Natural gas from Europe can be classified according to its calorific value: natural gases from the Netherlands have relatively low calorific values, whereas those from Russia usually have high calorific values. Moreover, countries have not yet agreed on a unified standard for the proportion of hydrogen to be injected into natural gas. The UK and Japan stipulate that the proportion of hydrogen in the natural gas pipeline network should be limited to less than 0.1%, while this ratio is 1% in Sweden and Belgium, 6% in France, and 10% in Germany (only for special cases) [6–9]. By contrast, natural gases in Europe are more tolerant of calorific values, which means a higher fraction of hydrogen is acceptable [10,11]. However, the standards applied in Japan are relatively strict. The authorities in Japan believe that the same proportion of hydrogen significantly impacts the HCNG quality, which cannot, therefore, meet the requirements [12–15].

According to the combustion characteristic index for HCNG calculated by Wang et al. [16], a hydrogen volume fraction of less than 27% (determined by the Wobbe Index and higher heating value (HHV)) had little effect on downstream customers. Jones et al. [17] gradually increased the injected hydrogen ratio of natural gas in natural gas appliance experiments until the flash-back limit, and found that a ratio of 34.7% was the upper limit of the injected hydrogen fraction, which occurred an obvious phenomenon of flash-back at extinction. Based on this, Zhao et al. [18] modified their experiments and believed it was safer and more reliable to set an upper limit of 25% of injected hydrogen fraction. By combining the interchangeability of HCNG and the combustion characteristics, Li et al. [19] proposed that 20–27% was a reasonable range of hydrogen fractions for injection into HCNG. Researchers from the UK pointed out that the maximum injected hydrogen ratio is 10% in natural gas pipeline transportation. This upper limit was 23% for domestic gas appliances and less than 5% for natural gas turbines. Researchers from Belgium calculated the Wobbe Index

and found that HCNG with a hydrogen ratio of less than 17% could be directly used in household/commercial stoves [20,21].

The following major problems have been encountered in studies of hydrogen mixing ratios for injection into natural gas pipelines:

- 1. Differences in gas quality are not considered in long-distance transportation and urban gas pipeline networks.
- 2. Natural gas is often analyzed as pure methane (CH_4) or fixed components, without considering the effects of impurities (C_2, C_3, N_2, CO_2) from the actual gas source.
- 3. Periodic changes in composition and calorific value of the same natural gas source are often not considered.

Without considering the impact of pipe material safety and other factors, this paper selects the real components of natural gas for half a year to one and a half years, according to the commissioning of various gas source stations in Zhejiang Province, China, by determining the relevant requirements for natural gas quality according to the current national standards. The calorific values and Wobbe Index values of hydrogen-blended gas for different hydrogen blending ratios were calculated, and the optimum range of hydrogen blending ratios, based on real gas sources, was studied from the perspective of natural gas quality.

2. Quality and Interchangeability Requirements for Natural Gas

In order to analyze the differences in the calorific values of HCNG following hydrogen admixing with different gas sources in Zhejiang Province, this section examines and analyzes the requirements of the relevant domestic standards.

2.1. Natural Gas Quality Requirements for Long-Distance Transportation

As shown in Table 1, natural gas falls into one of two categories in the national standard GB 17820-2018 (https://www.chinesestandard.net/PDF/English.aspx/GB178 20-2018, (accessed on 29 December 2021)) according to its HHV and the mole fractions of H₂S and CO₂ present in it. The HHV of Class I natural gas should be greater than 34 MJm⁻³, and that of Class II should have a HHV greater than 31.4 MJm⁻³. The natural gas transported in long-distance pipelines shall meet the quality requirements of Class I.

Category		Class I	Class II
Higher heating value $a,b/(MJm^{-3})$	\geq	34.0	31.4
Total sulfur ^a /(mgm ⁻³)	\leq	20	100
$H_2S^{a}/(mgm^{-3})$	\leq	6	20
Mole fraction of $CO_2/\%$	\leq	3.0	4.0

Table 1. Quality requirements for different types of natural gas.

^a The reference conditions used in this standard are 101.325 kPa, 20 °C. ^b Higher heating value was acquired on the base of dry basis.

Additionally, the national standard GB/T 37124-2018 provides more detailed requirements for natural gas quality in long-distance transportation pipeline networks. Table 2 further summarizes the upper limits of the mole fractions of CO, H₂, and O₂ permitted in pipelines, where the H₂ mole fraction should be less than or equal to 3%. Since this paper explores the proportion threshold of hydrogen injected into future natural gas pipelines, the limit of hydrogen concentration set by the standard is not considered temporarily.

Parameter		Value
Higher heating value ^{a,b} /(MJm ⁻³)	\geq	34.0
Total sulfura/(mgm ^{-3})	\leq	20
$H_2S^{a}/(mgm^{-3})$	\leq	6
CO ₂ mole fraction/%	\leq	3.0
CO mole fraction/%	\leq	0.1
H_2 mole fraction/%	\leq	3.0
O_2 mole fraction/%	\leq	0.1
Water dew point/°C	\leq	The water dew point should be 5 °C lower than the ambient temperature of transportation

Table 2. Gas quality requirements for natural gas in long-distance pipelines.

^a The reference conditions used in this standard are 101.325 kPa, 20 $^{\circ}$ C. ^b Higher heating value was acquired on the base of dry basis.

2.2. Natural Gas Quality Requirements of Urban Gas Pipelines

Based on the conditions mentioned in national standard GB 50028-2006 for urban gas transportation, its calorific value, composition, and water dew point should at least meet the requirements of Class II gas. In other words, urban gases should have HHV of greater than or equal to 31.4 MJm^{-3} , with a hydrogen content of less than 3%.

2.3. Wobbe Index for Long-Distance Natural Gas Pipeline Networks

The main parameters for studying natural gas interchangeability include a high Wobbe Index, the De Broglie exponent, the Weaver exponent, and HHV [22]. Of these, the Wobbe Index is an internationally accepted parameter for quantifying natural gas interchangeability. Therefore, this paper also uses this parameter to discuss the interchangeability of HCNG for different injected hydrogen ratios in the following sections.

As mentioned in the GB/T 37124-2018 standard, the Wobbe Index used to measure gas interchangeability in long-distance natural gas pipelines is recommended to range between 42.34 and 53.81 MJm⁻³. It is worth noting that the interchangeability mentioned above refers to the interchangeability between the alternative/mixed gas source and the existing gas source. Additionally, a 5% fluctuation in the Wobbe Index is considered reasonable, which is determined by the local historical average interchangeability of natural gas and that of the new gas source.

2.4. Wobbe Index in Urban Gas Pipelines

Urban natural gas should be classified according to a combination of the gas categories defined above and the Wobbe Index in order to control fluctuations in its calorific value accurately. According to the calorific value and Wobbe Index of natural gas, it can be divided into 3T, 4T, 10T, and 12T. The urban pipeline gas is generally required to meet the requirements of 10T or 12T, and the long-distance pipeline gas is required to meet the requirements of 12T. Since the existing national standards GB 50028, GB 17820, and GB/T 13,611 do not adopt a common reference value and the range of fluctuation of the Wobbe Index, the interchangeability of urban natural gas is referred to as the range given at 15 °C in the GB 13611-2018 standard. In addition, different classes of natural gas should follow the respective requirements of 10T or 12T, as listed in Table 3. Therefore, the Wobbe Index of Class I natural gas lies in the range $45.66-54.77 \text{ MJm}^{-3}$, while that of Class II natural gas lies in the range $39.06-44.84 \text{ MJm}^{-3}$.

Category		Wobbe Index/(MJm ⁻³)		
		Standard Value	Range	
Natural gas	10T	41.52	39.06-44.84	
	12T	50.72	45.66-54.77	

Table 3. Characteristic index of different classes of urban natural gas (at 15 °C and 101.325 kPa).

3. Case Study

Natural gas in Zhejiang Province has the characteristics of several gas sources, a complex pipeline network, large differences in the calorific values of different gas sources, and large fluctuations in the calorific values of different gas supply cycles. For example, there are seven natural gas sources currently operating in Zhejiang Province, which connect to 12 related gas stations. In the following sections, the calorific values of natural gas from seven different natural gas stations were examined and analyzed. According to the gas supply cycle and practical operating conditions of each gas station, the actual calorific value of natural gas was sampled and collected over a period of 0.5–1.5 years with a sampling frequency of once per hour.

3.1. HCNG Calorific Value and Wobbe Index Calculations

• Main parameters (Table 4)

Table 4. Density and high heating value (HHV) of natural gas components (in ideal gas volume).

Components	CH_4	C_2H_4	C_3H_6	$iso-C_4H_{10}$	<i>n</i> -C ₄ H ₁₀	iso-C ₅ H ₁₂
d (kgm ⁻³)	0.5548	1.0467	1.5496	2.0723	2.0787	2.48
HHV (MJm ⁻³)	37.044	64.91	92.29	119.28	119.66	146.76
Components	$n-C_5H_{12}$	C ₆ H ₁₄	N_2	CO ₂	H ₂	
d (kgm ⁻³)	2.6575	2.9	0.9671	1.5275	0.06953	
HHV (MJm ⁻³)	147.04	174.46	0	0	11.889	

• Calorific value of HCNG

$$H = \frac{1}{100}(H_1f_1 + H_2f_2 + H_3f_3 + \dots + H_nf_n) = \frac{1}{100}\sum_{r=1}^n H_rf_r$$
(1)

where *H* is the HHV of the natural gas (units of MJm⁻³), H_r is the HHV of combustible components in the natural gas (units of MJm⁻³), and f_r is the volume fraction of combustible components in the natural gas (units of %).

Relative density of HCNG

$$d = \frac{1}{100}(d_1f_1 + d_2f_2 + d_3f_3 + \dots + d_nf_n) = \frac{1}{100}\sum_{v=1}^n d_vf_v$$
(2)

where *d* is the relative density of the natural gas ($d_{air} = 1$), d_v is the relative density of combustible components in the natural gas, and f_v is the volume fraction of combustible components in the natural gas (units of %).

• Wobbe Index of HCNG

$$W = \frac{H}{\sqrt{d}} \tag{3}$$

where *W* is the Wobbe Index (units of MJm^{-3}), *H* is the HHV (units of MJm^{-3}), and *d* is the relative density of the natural gas.

As shown in Figure 2, the provincial pipeline network of natural gas in Zhejiang Province has a pattern of multiple gas sources and one ring network. The upstream is connected to eight gas sources, namely western first line, western second line, Sichuan gas, Donghai gas, Ningbo LNG, Zhoushan LNG, Xinjiang coal-to-gas, and Lishui 36-1. In the middle reaches, 2513 km of provincial long natural gas pipeline network has been built, with 91 stations and 109 valve chambers, which need to meet the requirement of a 12T gas supply. The downstream medium and low pressure pipeline is more than 30,000 km long, which needs to meet the gas supply requirement of 12T.



Figure 2. Zhejiang provincial pipeline network of natural gas.

• Main parameters

The light blue line in Figure 3a is the real calorific value of gas source station #1, the gas source for which is Ningbo LNG, and its characteristic is that the calorific value fluctuates between 36 and 39 MJm⁻³. Because of the large fluctuations in the green line within the gas supply period, this indicated that the gas quality was not sufficiently stable at station #1.

The pink line in Figure 3a is the calculated calorific value of the gas source mixed with hydrogen. When the hydrogen admixture reaches 10%, it reaches the boundary requirement of 34 MJm⁻³ for the gas intake calorific value of the long-distance pipeline. If it exceeds 10%, it fails to meet the long-distance pipeline intake requirements. The dark blue line in Figure 3a indicates when the hydrogen content reaches 20%, it reaches the urban gas calorific value boundary requirement of 31.4 MJm⁻³, and when it exceeds 20%, it will fail to meet the urban gas pipeline intake requirements.



Figure 3. (a) Calorific value of natural gas source station #1 measured between January 2020 and April 2020. (b) Wobbe Index of natural gas source station #1 measured between January 2020 and April 2020.

Figure 3b shows the Wobbe Index calculated based on the real gas source of gas source station #1. The range of variation of natural gas mixed with hydrogen below 20% is within the range of 12T natural gas, which meets the requirements for use.

The light blue line in Figure 4a is the real calorific value of gas source station #2, the gas source for which is Sichuan gas, with a calorific value between 36 and 37 MJm^{-3} most of the time. The range of fluctuation and amplitude of the calorific value of natural gas is relatively small with the gas supply time, and the gas quality is relatively stable. There is only some volatility in a fraction of the time. The pink line is the calculated calorific value of the gas source mixed with hydrogen. When the hydrogen admixture reaches 5% by content, the boundary requirement of the gas intake calorific value of the long-distance pipeline is 34 MJm^{-3} . If it exceeds 5%, it fails to meet the intake requirements of the long-distance pipeline. The dark blue line indicates when the hydrogen content reaches 16%, where it reaches the urban gas calorific value boundary requirement of 31.4 MJm^{-3} , and when it exceeds 16%, it fails to meet the urban gas pipeline intake requirements.



Figure 4. (a) Calorific value of natural gas source station #2 measured between January 2020 and April 2020. (b) Wobbe Index of natural gas source station #2 measured between January 2020 and April 2020.

Figure 4b shows the Wobbe Index calculated on the basis of the real gas source of gas source station #2. Except for a few days that only meet the gas supply requirements of 10T, the natural gas with the content of hydrogen doped less than 16% basically varies within the range of 12T natural gas, which meets the requirements for use.

The light blue line in Figure 5a is the real calorific value of gas source station #3. The gas source is the West–East Gas Pipeline. Its characteristic is that the calorific value fluctuates between 37 and 40 MJm⁻³, and the calorific value of natural gas fluctuates with the gas supply time. The range is very large, and the gas quality is too unstable. The pink line in Figure 5a is the calculated calorific value of the gas source mixed with hydrogen. When the hydrogen admixture reaches 12%, it reaches the boundary requirement of 34 MJm⁻³ for the gas intake calorific value of the long-distance pipeline. If it exceeds 12%, it fails to meet the long-distance pipeline intake requirements. The dark blue line in Figure 5a indicates when the hydrogen content reaches 23%, where it meets the urban gas calorific value boundary requirement of 31.4 MJm⁻³, and when it exceeds 23%, it fails to meet the urban gas pipeline intake requirements.



Figure 5. (a) Calorific value of natural gas source station #3 measured between March 2020 and March 2021. (b) Wobbe Index of natural gas source station #3 measured between March 2020 and March 2021.

Figure 5b shows the Wobbe Index calculated based on the real gas source of gas source station #3. The range of variation of natural gas mixed with hydrogen below 23% is within the range of 12T natural gas, which meets the requirements for use.

The light blue line in Figure 6a is the real calorific value of gas source station #4. The gas source is Sichuan gas. Its characteristic is that the calorific value fluctuates between $36.5 \text{ and } 37 \text{ MJm}^{-3}$, and the calorific value of the natural gas composition is very stable. The pink line is the calculated calorific value of the gas source mixed with hydrogen. When the hydrogen admixture reaches 10%, it reaches the boundary requirement of 34 MJm^{-3} for the gas intake calorific value of the long-distance pipeline. If it exceeds 10%, it fails to meet the long-distance pipeline intake requirements. The dark blue line in Figure 6a indicates when the hydrogen content reaches 20%, where it reaches the urban gas calorific value boundary requirement of 31.4 MJm^{-3} , and when it exceeds 20%, it fails to meet the urban gas pipeline intake requirements.



Figure 6. (a) Calorific value of natural gas source station #4 measured between August 2020 and March 2021. (b) Wobbe Index of natural gas source station #4 measured between August 2020 and March 2021.

Figure 6b shows the Wobbe Index calculated for the real gas source of gas source station #4. The range of variation of natural gas mixed with hydrogen below 20% is within the range of 12T natural gas, which meets the requirements for use.

The light blue line in Figure 7a is the real calorific value of gas source station #5. The gas source is East China Sea gas. Its characteristics are that the calorific value fluctuates between 37 and 40.2 MJm^{-3} and the ranges of fluctuation and amplitude of the calorific value of the natural gas vary with the gas supply time. They are very large and the gas quality is not sufficiently stable. The pink line in Figure 7a is the calculated calorific value of the gas source mixed with hydrogen. When the hydrogen admixture reaches 12%, it reaches the boundary requirement of 34 MJm^{-3} for the gas intake calorific value of the long-distance pipeline. If it exceeds 12%, it fails to meet the long-distance pipeline intake requirements. The dark blue line in Figure 7a indicates that when the hydrogen content reaches 22%, it meets the urban gas calorific value boundary requirements.





Figure 7b shows the Wobbe Index calculated for the real gas source of gas source station #5. Except for a few days that only meet the gas supply requirements of 10T, the

natural gas with the content of hydrogen doped less than 22% basically varies within the range of 12T natural gas, which meets the requirements for use.

The light blue line in Figure 8a is the real calorific value of gas source station #6. The gas source of gas source station #6 is West Second Gas. Its characteristic is that the calorific value lies between 37 and 38.5 MJm⁻³ most of the time, and the calorific value of the natural gas fluctuates with the gas supply time. The ranges of fluctuation and amplitude of the calorific value of the natural gas are relatively small, and the gas quality is relatively stable. There is some volatility only in certain months. The pink line in Figure 8a is the calculated calorific value of the gas source mixed with hydrogen. When the hydrogen admixture reaches 12%, it reaches the boundary requirement of 34 MJm⁻³ for the gas intake calorific value of the long-distance pipeline. If it exceeds 12%, it fails to meet the long-distance pipeline intake requirements. The dark blue line in Figure 8a indicates when the hydrogen content reaches 22%, where it meets the urban gas calorific value boundary requirement of 31.4 MJm⁻³, and when it exceeds 22%, it fails to meet the urban gas pipeline intake requirements.



Figure 8. (a) Calorific value of natural gas source station #6 measured between January 2020 and March 2021. (b) Wobbe Index of natural gas source station #6 measured between January 2020 and March 2021.

Figure 8b shows the Wobbe Index calculated for the real gas source of gas source station #6. The range of variation of natural gas mixed with hydrogen below 22% is within the range of 12T natural gas, which meets the requirements for use.

The light blue line in Figure 9a is the real calorific value of gas source station #7, the gas source for which is Lishui 36-1, its characteristic is that the calorific value lies between 35.5 and 36.5 MJm^{-3} most of the time, and the calorific value of the natural gas fluctuates with the gas supply time. The ranges of fluctuation and amplitude are relatively small, and the gas quality is relatively stable. There are fluctuations only on certain days. The pink line in Figure 9a is the calculated calorific value of the gas source mixed with hydrogen. When the hydrogen admixture reaches 6%, it reaches the boundary requirement of 34 MJm⁻³ for the gas intake calorific value of the long-distance pipeline. If it exceeds 6%, it fails to meet the intake requirements of the long-distance pipeline. The dark blue line in Figure 9a indicates when the hydrogen content reaches 17%, where it meets the urban gas calorific value boundary requirement of 31.4 MJm⁻³, and when it exceeds 17%, it fails to meet the urban gas pipeline intake requirements.



Figure 9. (a) Calorific value of natural gas source station #7 measured between January 2020 and March 2021. (b) Wobbe Index of natural gas source station #7 measured between January 2020 and March 2021.

Figure 9b shows the Wobbe Index calculated based on the real gas source of gas source station #7. The range of variation of natural gas mixed with hydrogen below 17% is within the range of 10T natural gas, which meets the requirements for use.

It can be seen from Table 5 that the variations in the hydrogen mixing ratios of different gas sources are quite different when the gas quality requirements are met.

Station	Hydrogen Doping Ratio (to Meet 34 MJm ⁻³)	Hydrogen Doping Ratio (to Meet 31.4 MJm ⁻³)
#1	$\leq 10\%$	≤20%
#2	\leq 5%	$\leq 16\%$
#3	$\leq 12\%$	≤23%
#4	$\leq 10\%$	$\leq 20\%$
#5	$\leq 12\%$	\leq 22%
#6	$\leq 12\%$	\leq 22%
#7	$\leq 6\%$	$\leq 17\%$

Table 5. Requirements for hydrogen mixing ratios of different gas source stations.

4. Conclusions and Suggestions for Future Research

It can be seen from the above analysis that the calorific values of different gas sources are quite different, and, at present, when studying the interchangeability of natural gas mixed with hydrogen gas in China, methane is basically analyzed as either a pure component or a fixed component. In order to calculate the calorific value of hydrogen admixing, Wobbe Index, etc., the obtained hydrogen mixing ratios do not represent the actual situation of real gas sources.

The pipeline network in Zhejiang Province is a complex network with multiple gas sources. There is mixed transportation of different gas sources in multiple pipe sections and at different times, so that the hydrogen mixing ratio cannot be fixed. Moreover, natural gas quality is closely related to particular gas sources and the gas supply cycle. When determining the threshold injected hydrogen ratio into natural gas, the gas quality of different sources and the supply period must both be considered. For example, according to the cycle characteristics of the gas supply, the method of dynamic hydrogen mixing ratio of fixed calorific value is to be adopted. Therefore, it is pointless to directly define a value/range for limiting the injected hydrogen ratios for HCNG, which should be formulated according to actual local conditions. **Author Contributions:** Conceptualization, X.L.; Methodology, S.T.; Investigation, S.T.; Resources, S.T.; Data curation, S.T.; Writing—original draft preparation, S.T.; Writing—review and editing, C.T., X.X.; Funding acquisition, X.L., S.S. and C.T. All authors have read and agreed to the published version of the manuscript.

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