

The Influence of increasing Hydrogen concentrations in the Natural Gas Supply Network

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ABSTRACT

With the necessity of new technologies to move energy systems from fossil to renewable sources, hydrogen takes an important role within the discussion. Whether in transport, industry, commerce, or private energy supply, hydrogen is predicted to be one of the main suppliers in our energy systems of the future. To enable a safe and easy solution for transportation, existing natural gas infrastructures can be used. Originally those structures were built for gas mixtures that contained higher amounts of hydrogen than the gas mixtures which are transported nowadays. But when fluctuating amounts of hydrogen are added to natural gas, it is important to determine the actual concentration of hydrogen - not only for the decreasing heating value but also the change in gas qualities and behavior of the mixture.

With advanced measurement principles and technologies, these concentrations could be determined due to their effect on thermodynamical properties. This paper summarizes the effects of hydrogen on natural gas mixtures and evaluates how these effects can be used to determine the composition of the gas. By knowing the hydrogen concentration, gas qualities can be recalculated and end devices adjusted to the changing conditions.

Keywords: hydrogen, renewable/green energy resources, energy storage, natural gas, climate change

NOMENCLATURE

Abbreviations

HNG	Hydrogen enriched Natural Gas
NG	Natural Gas
PtG	Power-to-Gas

Symbols

λ	Thermal conductivity [W/(m·K)]
μ_{JTCO}	Joule-Thomson Coefficient [K/MPa]
w_s	Speed of Sound [m/s]

1. INTRODUCTION

In order to reach climate goals and to decarbonize energy systems and infrastructures the feed-in of hydrogen into the natural gas supply network is a rational transition from fossil energy resources to renewable and environmentally friendly energy supplies.

While the European natural gas infrastructure is predicted to transport mainly synthetic methane and hydrogen in the future [1] it can be used already for the transportation of hydrogen to utilize excess electrical energy out of fluctuating resources. Due to technical limitations and the absence of mass-market-ready measurement technologies, the concentration of hydrogen in natural gas is strictly limited in most countries. Increasing hydrogen concentrations result in decreasing heating values, and therefore decreasing energy output while burning the same volume of HNG compared to NG.

Different studies (e.g. by DVGW [2]) proved the reliable work of several devices with increasing hydrogen concentrations multiple times. Nevertheless, the gas quality changes significantly with increasing hydrogen concentration. Therefore, the actual hydrogen

concentration needs to be known. While compositions of NG in the gas supply network vary because different sources feed in different gas qualities, the determination of the composition requires an input of various measured data points. Not only the hydrogen concentration needs to be determined-different independent parameters need to be measured to deduce the composition.

2. MOTIVATION AND APPROACH

For a summary of possible solutions and some additional background information, the following sections contain the foundation of the Power-to-X, respectively Power-to-Gas approach, as well as the relevant parameters and properties, which evaluate gas qualities. Furthermore, a short survey of relevant thermodynamical properties is given.

2.1 Power-to-Gas

There are many different approaches to improve and develop energy infrastructure and to increase efficiencies. The conversion of electrical energy into different products as for example heat (Power-to-Heat), liquids (Power-to-liquid), or gas (Power-to-Gas) has the potentials to make use of excess electrical energy. PtG implies the separation of water via electrolysis. The fluctuating energy sources wind and solar power can

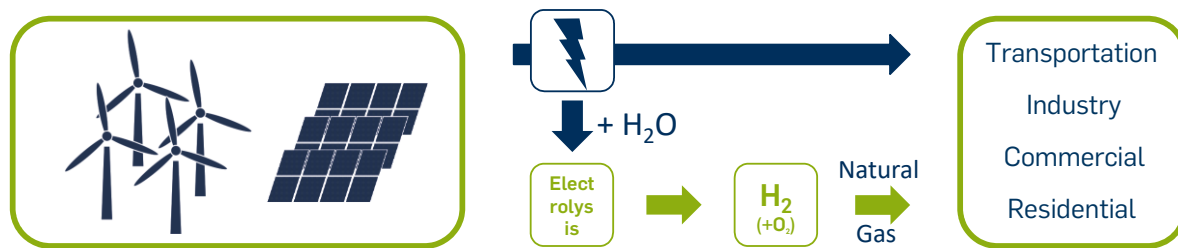


Fig. 1. Concept of PtG to increase efficiency of fluctuating renewable energy resources like wind and solar power

therefore be transformed into (H₂+O₂) but without an existing hydrogen infrastructure transportation requires additional energy due to specific pressure and temperature levels.

Some approaches provide further conversions into methane (synthetic NG) or ammonia to solve the question of transportation. Since the losses increase with more conversion steps the direct use and transportation of the (green) hydrogen is very efficient.

2.2 Gas qualities

To ensure constant standards and qualities despite the varying composition, different properties are measured and tracked in the supply network constantly. Depending on countries and their limitations for different consumers, properties as the higher/lower heating value, the relative density, the methane number as well as the Wobbe-Index are used for characterizing the qualities of NG.

Especially the decreasing heating value and relative oxygen demand result in dropping efficiencies and therefore a lower energy output. Enrichments of 20 mol-% H₂ result in a decrease of heating value by +13 % in total. While the specific heating value of hydrogen is very high, the volumetric heating value is only approximately 30 % compared to pure methane. This cannot only lead to pressure gradients in the supply network but also damages devices that are adjusted for specific operation points. Furthermore, a decreasing relative oxygen demand results in a loss of efficiency due to preheating more air than necessary for combustion processes.

Conventionally the composition and therefore the gas qualities are measured with technologies that are based on infrared spectroscopy or chromatographic devices which extend a small inline measurement system by far.

2.3 Thermodynamical properties

Besides the impact on gas qualities as mentioned in the chapter above, increasing hydrogen concentrations lead to significant changes in thermodynamical properties as well. These changes of properties do not directly influence the demanded qualities, but can be used to determine the concentrations which are the missing information to operate safely and reliably with specific hydrogen enrichment.

Properties such as density, speed of sound, and isobaric/isochoric heat capacity, as well as transport properties such as thermal conductivity, viscosity, and

dielectric constant of gas mixtures show high sensitivity for increasing hydrogen concentrations. However, each component contributes to a change in the behavior of the mixture. The circumstance of varying compositions of the natural gas complicates the determination of the hydrogen concentration if not at least the composition right before a hydrogen feed-in is known. While varying concentrations of the main components of NG (hydrocarbons), only change the gas qualities within the tolerances, changing hydrogen concentrations have immense effects on the NG mixtures.

3. DETERMINATION OF H₂-CONCENTRATIONS

The theoretical approach which is used for the determination will be presented briefly in this section as well as challenges and further approaches.

3.1 Proceeding and Conceptualization

To take advantage of the sensitive parameters, as mentioned in the section above, a model to determine hydrogen concentrations is developed. Therefore, the sensitivities of those parameters were analyzed and combined in different groups of possible solutions. The combinations which turned out to be most promising are further investigated with regard to their experimental realization.

To reduce the number of different properties that are necessary for the determination, properties in at least two different states are considered. Therefore, the real gas behavior of the different components and their effect on the mixture in total can be used. And while pressure gradients of at least 2-5 MPa are necessary to make use of the residual behavior of properties, the change of temperature while throttling the pressure can be related to the Joule-Thomson Coefficient (μ_{JTCo}) of the gas mixture. This coefficient is another property, which is highly influenced by hydrogen concentrations. While the main part of NG, the hydrocarbons tends to cool down while isenthalpic throttling, hydrogen reacts the opposite way due to the negative value of μ_{JTCo,H_2} .

To calculate the hydrogen concentration of a multicomponent mixture as NG an iterative method is used. The potentially measured properties (e.g. speed of sound, w_s) are fed into an algorithm that compares those values to a modeled gas composition. These properties are calculated with the software TREND [5] and the GERG-2008 [6] equation of state. Transport properties like thermal conductivity (λ) are calculated with the extended corresponding states model [7] implemented in REFPROP [8].

With a nonlinear algorithm, the sum of squared deviations between both compositions is constantly minimized. Based on a binary (CH₄ + H₂) mixture with the two main components of HNG, the calculation starts with polynomial functions. Therefore, coefficients are set for a specific pressure and temperature range to define wide limits for the following iterations to minimize the sum of squared deviations.

With each loop, one component is added to the modeled mixture and the sum of squared deviations is minimized iteratively again. Due to their high sensitivity, the thermal conductivity λ and the speed of sound w_s of HNG compositions turned out to be the most promising choices for thermodynamic parameters.

3.2 Results

In the following figure, the results of 140 calculations are represented. With an aim at absolute deviation between calculated and experimental concentrations of $\pm 2\%$ maximum, the five relevant components (methane, ethane, propane, nitrogen, hydrogen) are presented.

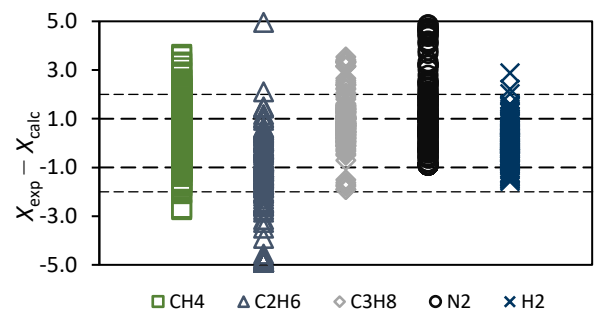


Fig. 2. Total deviations of calculated and exemplary composition presented for the main components

While the deviations for hydrogen fractions are mostly in between $\pm 2\%$, deviations of the other components scatter a lot more. Nearly 75% of the results of hydrogen concentrations are between +1 to -1 deviation in total. The smaller the fraction of a component, the less they influence the results. This is the reason why C₄₊ and CO₂ are not depicted in the figure. Deviations of C₄₊ scatter within a range of $\pm 1\%$ in total and deviations of CO₂ result in -1 to -2% in total. These components have main effects on the thermodynamical properties of the gas mixture and increasing concentrations would be detected immediately. This is why the deviations are as low as mentioned above.

3.3 Discussion and Conclusion

The main weakness of a calculation based on correlations and the iterative procedure is the scatter of results as presented in Fig. 2. Although nearly all hydrogen fractions are calculated within $\pm 2\%$ deviation in total, even ca. 75 % within $\pm 1\%$, scatter of the concentration of the remaining components is too high to ensure a reliable calculation of relevant gas qualities. Furthermore, the combined measurement uncertainties of each property (λ , w_s , p , T) are roughly estimated and the pseudo experimental values were impinged with the ranges of these uncertainties with a normal distribution. Their influence on the experimental setup combined with varying compositions may exceed the deviations which are calculated.

To reduce the scatter of the concentrations of all components, but especially the deviations of hydrogen fractions, additional properties need to be implemented into the model. Therefore, spectroscopic data (infrared spectroscopy) will be used to support the reliability of the calculation. Although hydrogen, respectively its absorption, cannot be detected by infrared absorption, it can be used to specify limits for the concentration of hydrocarbons and CO₂. This additional approach as well as the theory based on different thermodynamical properties will be investigated with an experimental setup that is currently under construction.

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